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# REE Recovery from End-of-Life NdFeB Permanent Magnet Scrap: A Critical Review

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**Abstract** NdFeB permanent magnets have different life cycles, depending on the applications: from as short as 2–3 years in consumer electronics to 20–30 years in wind turbines. The size of the magnets ranges from less than 1 g in small consumer electronics to about 1 kg in electric vehicles (EVs) and hybrid and electric vehicles (HEVs), and can be as large as 1000–2000 kg in the generators of modern wind turbines. NdFeB permanent magnets contain about 31–32 wt% of rare-earth elements (REEs). Recycling of REEs contained in this type of magnets from the End-of-Life (EOL) products will play an important and complementary role in the total supply of REEs in the future. However, collection and recovery of the magnets from small consumer electronics imposes great social and technological challenges. This paper gives an overview of the sources of NdFeB permanent magnets related to their applications, followed by a summary of the various available technologies to recover the REEs from these magnets, including physical processing and separation, direct alloy production, and metallurgical extraction and recovery. At

present, no commercial operation has been identified for recycling the EOL NdFeB permanent magnets and the recovery of the associated REE content. Most of the processing methods are still at various research and development stages. It is estimated that in the coming 10–15 years, the recycled REEs from EOL permanent magnets will play a significant role in the total REE supply in the magnet sector, provided that efficient technologies will be developed and implemented in practice.

**Keywords** Critical raw materials · Neodymium · Rare earths · Rare-earth magnets · Recycling · Urban mining

## Introduction

Neodymium–iron–boron ( $\text{Nd}_2\text{Fe}_{14}\text{B}$ , or NdFeB for short) permanent magnets are considered as the best available magnets since their introduction on the market in 1984, due to their superior energy product (with a theoretical maximum of  $512 \text{ kJ/m}^3$ ), which makes them highly efficient and suitable for lightweight mobile applications [1].

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Consequently, they are widely used in wind turbines, hybrid and electric vehicles (HEVs and EVs), household electrical appliances, computer hard disk drives (HDDs), and many small consumer electronic devices.

NdFeB permanent magnets have different life cycles, depending on the applications: from as short as 2–3 years in consumer electronics to 20–30 years in wind turbines. The weights of the magnets range from less than 1 g in small consumer electronics to over 1 kg in EVs and HEVs, and can be as large as 1000–2000 kg in the generators of modern wind turbines. NdFeB magnet contains about 31–32 wt% rare-earth elements (REEs), mainly Nd + Pr plus a few minor, but expensive, heavy rare-earth elements (HREEs) such as Dy, Tb, and Gd, depending on the applications. Recycling of REEs contained in this type of magnets from the End-of-Life (EOL) products will play an important and complementary role for the primary supply of REEs in the future. However, collection and recovery of the magnets from small consumer electronics imposes great social and technological challenges. At present, no mature and economically feasible technologies have been identified for recycling the EOL NdFeB permanent magnets and the recovery of the associated REEs. Most of the processing methods are still at the research and development stage. It is estimated that from 2030 on, the recycled REEs from the EOL permanent magnets will play a significant role in the total REE supply in the magnet sector [2]. Moreover, recycling of REEs is also of importance for the so-called Balance Problem, since it avoids producing excesses of La or Ce while mining of REE ores for Nd and/or Dy production [3–5].

To date, no commercial process to recycle these REEs from EOL products is operational. It is simply cheaper to buy REE master alloys or newly manufactured magnets originated from primary resources than to reprocess the complex scrap material from recycled resources [6, 7], especially for small-sized products where manual separation of the magnets is challenging. Moreover, since most household electrical and electronic devices are currently shredded, their magnetic content, mainly containing iron and neodymium, tends to end up in the ferrous metal waste stream, which is too diluted for economically feasible REE recovery.

There are different metallurgical processes to recover the REEs in the NdFeB magnet scrap at various technology-readiness levels (TRLs) [8–11], including hydrogen decrepitation [12, 13], chemical vapor transport [14, 15], liquid metal extraction [16], hydrometallurgical processing [17, 18], and pyrometallurgical slag extraction [19], among many others. However, the majority of these techniques were developed to recover the REEs from pre-consumer (manufacturing) scrap that is relatively clean and homogenous, and particularly having a high REE

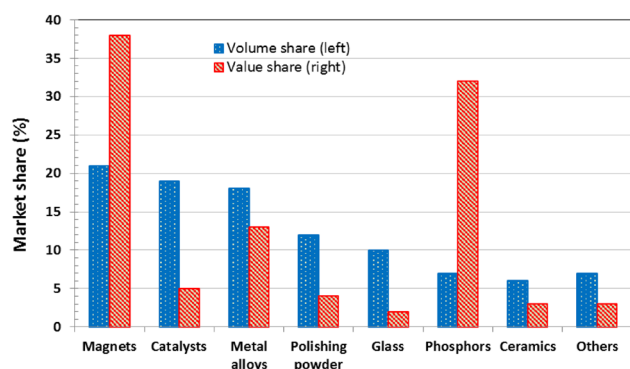
concentration. The recovery of REEs from complex post-consumer magnet scrap has not been well studied before. This paper gives an overview on the sources of EOL permanent magnets and characteristics of their occurrence, technical challenges and potential physical and metallurgical separations for the recovery of the magnets and the associated REEs, and finally a life cycle assessment (LCA) for recycling EOL NdFeB magnet scrap.

## Applications of NdFeB Magnets and the Source for Future Recycling

### NdFeB Magnet Production and Market Demand

The global annual REE production remained rather constant at about 120,000 tons (counted as oxides-REOs) during 2005–2015, except for two peak years of 2006 and 2009 (~137,000 tons) according to the USGS Minerals Yearbook for Rare Earths published annually during 2010–2016 [20]. Although very little public statistics are available for individual REE production in the past years, a thorough analysis of the global distribution of rare-earth oxide consumption by market sectors was reported for the year 2008 by the U.S. Department of the Interior and the USGS [21]. Approximately 26,000 tons of REEs (counted as REOs) are used per year in NdFeB permanent magnets, which form the largest applications among all REEs both in tonnage and in market value [22, 23], as shown in Fig. 1. However, it is very difficult to get accurate and reliable REE production figures for the use of the NdFeB magnets in the different applications, and thus many statistics are based on estimations from different sources and may differ significantly from one source to the other.

The great versatility of the REE magnet applications is also contributed by the fact that fine-tuning their chemical composition by adding some Dy, Tb, Gd, Nb, Co, Cu, Ga, and Al can modify some of the physical and magnetic properties to suit wide application needs [24]. Dy and Tb are used to increase the anisotropy and the coercivity of the magnet, which is of importance for magnets to be used at higher temperatures, but they also decrease the remanence and the energy product. Addition of Gd improves the temperature coefficient. Cu and Al are added to improve sintering of the magnet alloy, while Nb is added for grain refining. Ga improves the intrinsic coercivity and the hot workability of the alloy. Co is added to increase the Curie temperature, and some NdFeB magnets could contain up to 5 wt% Co. In 2008, about 26,300 tons of REEs including Nd, Pr, Dy, Gd, and Tb, counted as oxides, were used for magnet production globally. For the 26,300 tons of REOs above, about 76 % of neodymium (18,200 vs. 23,900 tons), 70 % praseodymium (6140 vs. 8740 tons), 69 %

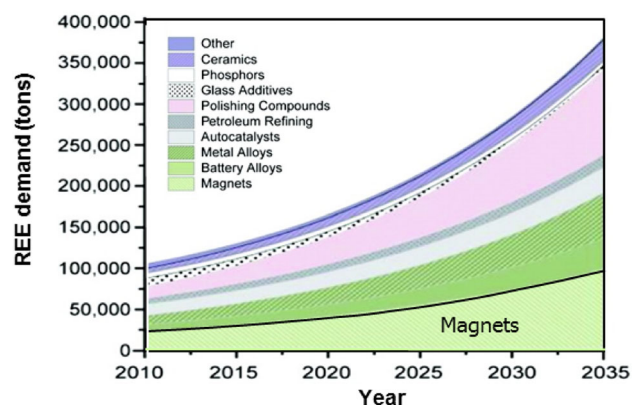


**Fig. 1** Market shares of REE magnets in terms of volume and monetary value (created based on data of BGS [22])

gadolinium (525 vs. 762 tons), and 100 % dysprosium (1310 tons) have been used for permanent magnet production in 2008 [21].

Figure 2 shows the predicted growths of all REEs according to the application areas [25]. As seen from the figure, the REE magnets and dysprosium will be the most demanded materials, largely due to the fast growing green technologies for clean energy and transportation (wind turbines and electric vehicles).

REE permanent magnet contains about 31–32 wt% REEs (mainly 21–31 wt% (Nd + Pr), 0–10 wt % Dy plus small amounts of Gd and Tb) [26]. Dysprosium is essential for high-temperature applications such as in EVs/HEVs. It is expected that the NdFeB permanent magnet market will continue to grow, and Fig. 3 illustrates the recent global NdFeB permanent magnet production and forecast to 2017/2020 [24], and Table 1 lists the magnet production in year 2012 and prediction for the year 2015 [27]. As clearly illustrated in Fig. 3 and Table 1, Japan and China have been the two main REE magnet-producing countries, and in recent years, China has been by far the dominant magnet-producing country with about 80 % market share. According to the 2013 forecast in the UNEP report, the



**Fig. 2** Predicted growth of REE demand (2010–2035) Adapted from Alnos et al. [25] (reprinted with permission)

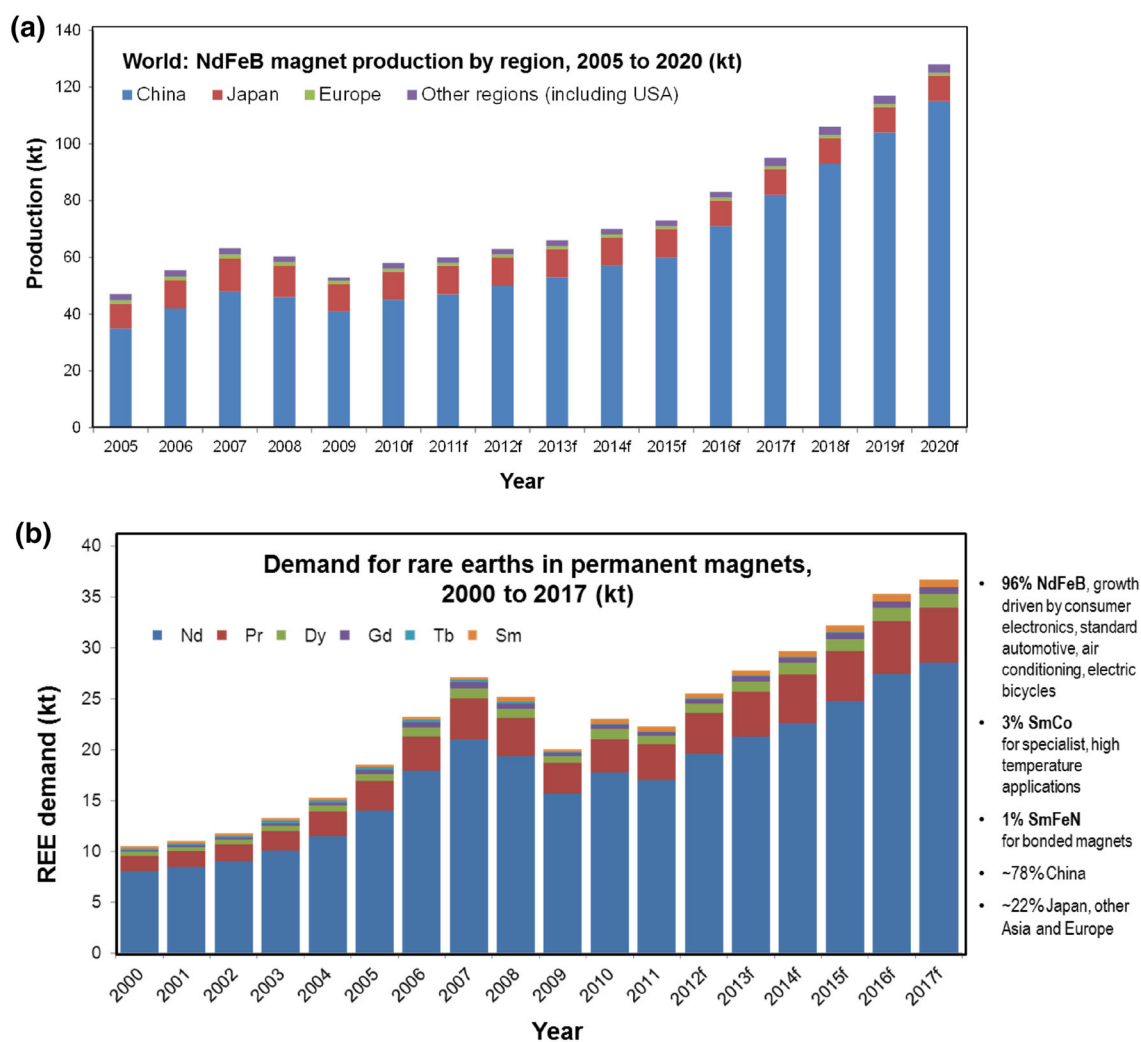
REO demand for REE magnet production will reach 36,000 tons, an equivalent of NdFeB magnet of approximately 99,500 tons [28]. However, the predicted figures for 2015 and 2016 may differ from the real demand and production significantly due to the weakened market demand in the recent years. Unfortunately, there exist no public statistics on the total NdFeB permanent magnet production and demand for the recent years to validate the former predictions. The known applications using NdFeB magnet (incomplete) are likely to come down showing a much lower figure as will be discussed in “Applications of NdFeB Magnets” section.

## Manufacturing and Applications of NdFeB Magnets

### *Properties and Production Methods of Rare-Earth Permanent Magnet (REPM)*

Today, key drivers for the development of new permanent magnet materials are the growing demands for industrial motors and automation, consumer electronics, e-mobility, and the shift toward renewable energies, most importantly wind power [29]. As a consequence of the 2010/2011 Rare Earth Crisis, research has also been driven by the quest for permanent magnet materials without [30] or with a reduced amount of critical rare-earth metals [31]. There is a paradigm shift in how this research problem is approached: while so far the impressive advances in permanent magnet research have almost exclusively been pushed by experimental materials science research, today, the modeling of new materials systems gains more importance [32].

Making a permanent magnet is governed by following two major principles: first, choosing the right starting material that comprises good intrinsic properties, that is, saturation magnetization, Curie temperature, and magnetocrystalline anisotropy; second, translating these intrinsic properties into functional extrinsic properties, most importantly a high remanent magnetization and coercivity (hysteresis). This is done by processing the materials appropriately and adjusting respective microstructures [33]; a schematic of main processing routes can be found in Fig. 4. In the case of Nd–Fe–B, the market is dominated by sintered, the so-called nucleation-type magnets. In this case, a Nd–Fe–B starting alloy is produced by a strip caster, which is used to cast the alloy on a water-cooled, rotating wheel that rapidly quenches the alloy. The great advantage of this technique is that the resulting alloy comprises a very homogenous microstructure with Nd–Fe–B grains in the microscale; the segregation of  $\alpha$ -Fe is largely avoided. In a second step, the material is pulverized by hydrogen decrepitation (HD) and then jet-milled down to a grain size of typically 3–5  $\mu\text{m}$  [34]. The powder is then aligned in a magnetic field and pressed to make a textured



**Fig. 3** Global production of NdFeB permanent magnets and the demand for the REEs [23] (reprinted with permission). **a** Total global NdFeB magnet production and prediction: 2005–2020, **b** Total global REE demands for permanent magnets

**Table 1** Production of NdFeB permanent magnets ( $\times 1000$  metric tons) [27]

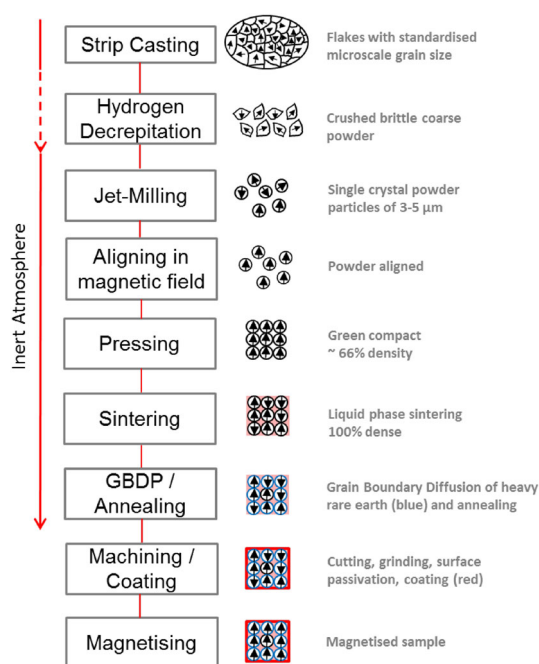
| Production by country/region                                | 2012 | 2015 forecast |
|---|------|---------------|
| China   | 50   | 65            |
| Japan   | 10   | 8             |
| Europe  | 1    | 1             |
| USA   | 0    | 2             |
| All others  | 2    | 2             |
| Total REE magnet  | 63   | 78            |
| Total REE in magnet (31 wt%), estimates                     | 19.5 | 24.2          |
| Total REO equivalent (in terms of $\text{Nd}_2\text{O}_3$ ) | 22.8 | 28.2          |

green compact, which is then sintered. The overall REE content of the magnet is considerably higher as suggested by the stoichiometry of the main phase: the extra REE

portion makes it possible to run a liquid-phase sintering process. The sintered body is then finally annealed to improve the magnetic properties, most importantly the coercivity. The REE-rich, intergranular low-melting eutectic phases are the key to enhance coercivity, that is, the ability of the magnet to withstand demagnetization: the idea is to magnetically decouple the grains in order to avoid the movement of transverse domains as much as possible. The magnets are mechanically worked (sawing, grinding, polishing) to get a final product. In terms of (raw materials) cost, this final production step should not be underestimated: many tons of REE-rich grinding slurry are accumulated each year, a valuable resource, in respect of which an appropriate, cost-effective, and clean recycling process is still a challenge.

Magnetization reversal, i.e., the nucleation of domains after the magnet has first been saturated and mounted and then been exposed to an opposing magnetic field in the





**Fig. 4** Schematic of modern manufacturing process of sintered NdFeB magnet

application, happens by nucleation at “imperfect” interfaces of individual Nd–Fe–B grains (“nucleation-type magnet”)—“imperfect” in the sense of magnetic, structural, chemical, or geometrical imperfections [35]. This observation led to the development of the so-called grain boundary diffusion process, in which HREEs, most importantly Dy and Tb, are diffused into the microstructure of the magnet along the grain boundaries, magnetically hardening particularly these areas of the grains (Dy and Tb comprise higher magnetocrystalline anisotropies than Nd). Apart from the cost-wise benefits, the reduced addition of HREEs increases the remanence of highly coercive Nd–Fe–B, thus the energy density,  $(BH)_{\max}$ , due to the lower amounts of HREE 4f spins that couple antiparallel with the 3d electrons. Interestingly, this kind of advanced processing experienced its industrial breakthrough in the aftermath of the 2010/2011 global Rare Earth Crisis, when prices particularly for HREE oxides were multiplying by several orders of magnitude.

The second major class of Nd–Fe–B permanent magnets are nanocrystalline, polymer-bonded magnets. The main steps of the production route adopted are (1) rapidly quenching a Nd–Fe–B alloy by melt spinning, that is, at much higher solidification speeds than those achieved by a strip caster, to produce polycrystalline flakes with grains in the nanoscale; (2) milling these flakes to a standardized particle size; and (3) mixing the flakes with a polymer and injecting or pressing this compound into a magnet. The major advantage of this approach is the near-net-shaped production of magnets with more complex geometries.

Energy densities of these kinds of magnets are, of course, much lower than those of sintered magnets: the permanent magnet material is “diluted” by the polymer, and the standard bonded magnets are isotropic, which means that the Nd–Fe–B grains in the polycrystalline particles are not textured.

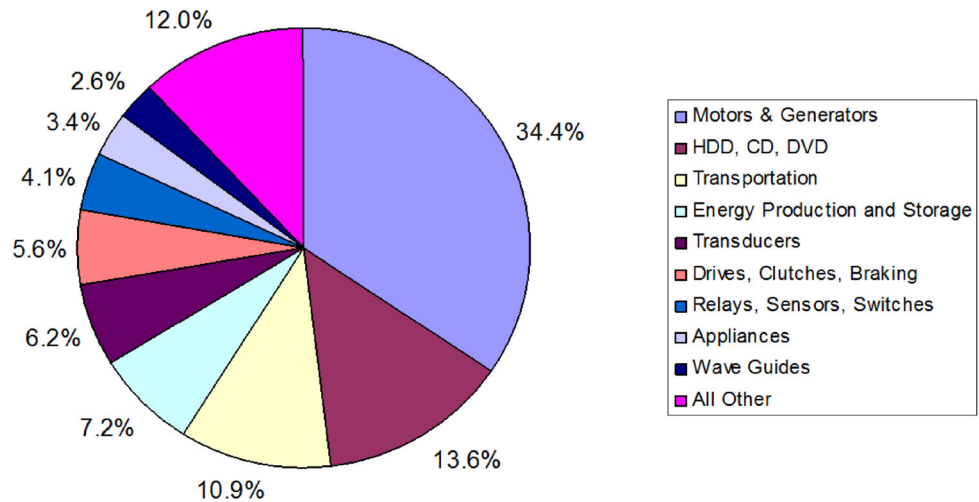
There are two ways to induce texture in a polymer-bonded magnet: (1) using melt-spun flake materials that underwent a hot deformation process (see below); or (2) using materials processed in a dynamic hydrogenation disproportionation desorption and recombination (d-HDDR) process [36]. In the d-HDDR process, larger single-grain particles obtained by strip casting and hydrogen decrepitation (see above) are disproportionated at 800–900 °C hydrogen pressures of up to 100 kPa to form anisotropic, polycrystalline particles in the nanoscale. The dynamic control of hydrogen pressure and temperature during the process makes it possible to preserve a high degree of texture within the individual grains—the so-called texture memory effect [37]. Finally, it is important to note that HD and HDDR processing constitutes an important recycling pathway for NdFeB magnets [38–41].

Nanocrystalline, melt-spun Nd–Fe–B can also be hot pressed and hot deformed to make isotropic and anisotropic permanent magnets, respectively [42]. By backward extrusion of the compacts, ring-shaped magnets or those with a high degree of texture and radial magnetization can be produced [43]. The great advantage of nanocrystallinity are the high values of coercivity that can be achieved, since grain size correlates negatively with coercivity until the sizes of the grains scale-up to match with those of the critical single-domain size of Nd–Fe–B, that is, at around 300 nm. In addition, nanocrystalline materials are more corrosion resistant than sintered magnets [44]. The disadvantages of hot pressing and compaction are the relatively high processing costs, caused by the noncontinuous nature of the batch-process and expensive pressing tool that have to be changed regularly. In addition, energy densities of hot-deformed magnets are lower than those of sintered magnets, due to a lower degree of texture that can be achieved.

#### *Applications of NdFeB Magnets*

According to Constantinides [45], more than 20 types of applications could be identified for rare-earth magnets. Different ways of classifications for REE magnet applications can be found. Figure 5 shows the relative share of REE magnet applications, predicted for the 2012 year end by Shaw and Constantinides [23]. HDDs and CD/DVDs used to be the number 1 applications of the REE permanent magnets, and as is seen from the data, motors and generators are, however, becoming the most important

**Fig. 5** Shares of the different applications in the global NdFeB market for year 2012 [23] (reprinted with permission)



**Table 2** Applications of NdFeB permanent magnets and the estimated annual demand for 2014/2015

| Sectors              | Product                                   | Concrete use                       | Mass per unit  | Total use (ton)     | Remarks/References   |
|----------------------|---|------------------------------------|----------------|---------------------|--|
| Computers            | HDDs (excluding CDs, DVDs)                | VCM, SP motors                     | 10–20 g        | 7500 (2015)         | Constantinides in 2016 [45]  |
| Consumer electronics | Home electronics & electrical appliances* | Small electric motors              | Varies greatly | No total statistics | Speakers in sound system, mobile phones and music players; washing machines, refrigerators etc.  |
|                      | – Air conditioners                        | 3–4 motors                         |                | >4000 (2014)        | Constantinides in 2016 [45]  |
|                      | – Speakers                                | From mobile phones to cars         |                | >4500 (2015)        | Constantinides in 2016 [45]  |
| Wind turbines        | Generation IV (only)                      | Generators                         | 400 kg/MW      | 8500 (2015)         | Constantinides in 2016 [45]  |
| Vehicles             | Conventional automobiles                  | Small motors (40) and sensors (20) | 250 g          | ~ 22,000 (2015)     | Estimates based on Shaw and Constantinides in 2012 [23] and Badanra et al. [46] for magnet/car, and about 89.7 million car sales in 2015 [47]                            |
|                      | HEVs, PEHVs and EVs (average)             | Electric motors                    | 1.25 kg**      | >7000 (2015)        | For hybrid Electric Cars and Trucks. Constantinides in 2016 [45]   |
|                      | Electric bikes (E-bikes)                  | Electric motors                    | 300–350 g      | 6000 (2015)         | 35 million units/year predicted for 2015 by Constantinides in 2012 [48], adding up to ~ 10,000 tons. Updated figure of 6000 tons for 2015 by Constantinides in 2016 [45] |
| Total (incomplete)   |   |                                    |                | 51,000 (100 %)      | This figure does not include the magnet consumption from major household appliances and CD/DVD drives, but air conditioners are included                                 |

\* Large household appliances include refrigerators (containing 40–60 g NdFeB magnets per unit), washing machines (80–180 g NdFeB/unit) [46]. Suppose an average REE magnet of 50 g/unit and a global sales of 200 million unit per year: total magnet consumption will be 10,000–20,000 ton/year for this sector

\*\* This figure does not include the magnets in the small motors and sensors like in the conventional cars

applications in recent years. Although these figures are relatively old, and there are quite some discrepancies from different sources and for different years, it is nevertheless possible to observe some general trends. The classification and grouping of applications are sometimes different, and will lead to data inconsistencies. It is now a commonly

accepted fact that the market for HDD and CD/DVD drives is shrinking, and that the use of the magnets in the transport sector, in particular EV/HEV and E-bikes, is rapidly growing.

Table 2 lists the main applications of NdFeB magnets and the relative market size and the total estimated annual

demand. The total estimated demand for the REE magnets is about 63,000 tons, excluding the household appliances. According to the above incomplete statistics for 2015 in [45], the main application sectors are conventional automobiles (43 %), wind turbines (17 %), computer HDDs (15 %), EVs/HEVs (14 %), and electric bikes (12 %). However, large uncertainties arise from the use of magnet in conventional vehicles. If only the reported incomplete figures for 2015 by Constantinides [45] are used, the total reported six sectors accounted for 37,500 tons, and the shares of the six reported sectors will be wind (23 %), HDD (20 %), HEV/EV (19 %), e-bikes (16 %), speakers (12 %), and air conditioners (10 %). When recycling is concerned, the wide range of life span of different products needs to be carefully considered.

### Recycling Potential of NdFeB Permanent Magnets

According to the estimations of Du and Graedel [49], global in-use stocks for four REEs in NdFeB permanent magnets, i.e., neodymium (Nd), praseodymium (Pr), terbium (Tb), and dysprosium (Dy), amount to a total of 97.0 kilotons (kt): 62.6 kt Nd, 15.7 kt Pr, 15.7 kt Dy, and 3.1 kt Tb. These stocks, if efficiently recycled, could provide a valuable supplement to geological stocks as they are almost four times the 2007 annual extraction rate of the individual elements. Different applications of the four REEs as permanent magnets estimated by Du and Graedel [49] are listed in Table 3.

Rademaker et al. [2] estimated the recycling potential of NdFeB permanent magnets for three application sectors by considering the manufacturing and life span of the applications: computer hard disk drives, wind turbines, and automotive industry. The results show that for some time to come, waste flows from permanent magnets will remain small relative to the rapidly growing global REE demand. Policy makers therefore need to be aware that during the next decade, recycling is unlikely to substantially contribute to the global REE supply. In the long term, waste flows will increase sharply and will meet a substantial part of the total demand for these metals. Figure 6 shows the recycling potential of Nd and Dy for the three studied application sectors. Future REE recycling efforts should, therefore, focus on the development of recycling technology and infrastructure. Those authors also emphasized that one of the main challenges for recycling of NdFeB magnets from consumer products is the physical dismantling and up-concentration of small NdFeB magnets in diversified scrap. REE magnets in wind turbines and EV/HEV vehicles are more easily dismantled and physically concentrated, and even reuse of these large magnets is possible after refurbishing. In general, efficient metallurgical separation and refining processes remain the main challenges. It

is estimated that approximately 5–10-year period is required to set up a recycling practice.

Guyonnet et al. [50] reported their research on the material flow analysis as applied to REEs in Europe. Based on the analysis of Nd and Dy metal in magnet applications in 2010 in the EU, 1230 tons of Nd flows into the EU to be used for NdFeB magnets, among which at least 597 tons of Nd has been reported into the waste stream (207 tons as landfill and dissipative loss, and 390 tons dispersed in steelmaking) with zero recycling. They indicated that projections for recycled flows around year 2020 for Nd in permanent magnets are on the order of 170–230 tons per year in the EU.

The study of Habib et al. [51] reveals that a normal primary supply is unable to meet the forecasted demand of Nd and Dy in their modeled demand scenarios by 2050. Although recycling is unlikely to close the wide gap between future demand and supply by 2050, in the long term, secondary supply from recycling can meet almost 50 % of the demand, i.e., by 2100. It is evident that recycling can play a major role in reducing the geopolitical aspects of supply risk.

### Physical Separation Technologies

#### Destination of the Permanent Magnets in Different EOL Products

Except for generators and motors in wind turbines and EV/HEVs, the REE permanent magnets are not pre-dismantled, due to their relative small size, from their attached EOL products in computer HDDs and other home electronics such as mobile phones, music players and speakers in the sound systems. The EOL electronic products (e-waste) are generally shredded as a whole after removal of the hazardous components/materials. Due to the strong permanent magnetism, the small magnets will stick to ferrous scrap (steel) after shredding and physical separation using such as magnetic separators, or to a much smaller extent trapped to the nonferrous scrap if the magnets are not liberated and still stuck to the nonferrous components. At present, almost all small permanent magnets used in consumer electronics (at relatively low collection rate), after collection and shredding operation, are lost into ferrous or nonferrous scrap and reported into slag or residues during pyrometallurgical or hydrometallurgical recovery processes of the major metals, which was recently proved by the study of Bandara et al. [52] from the sampled Electric Arc Furnace (EAF) steelmaking slag. EAF steelmaking uses almost 100 % steel scrap as raw materials which may come from various secondary resource, including shredding of EOL vehicles (ELVs) and electronic waste. Their analysis of



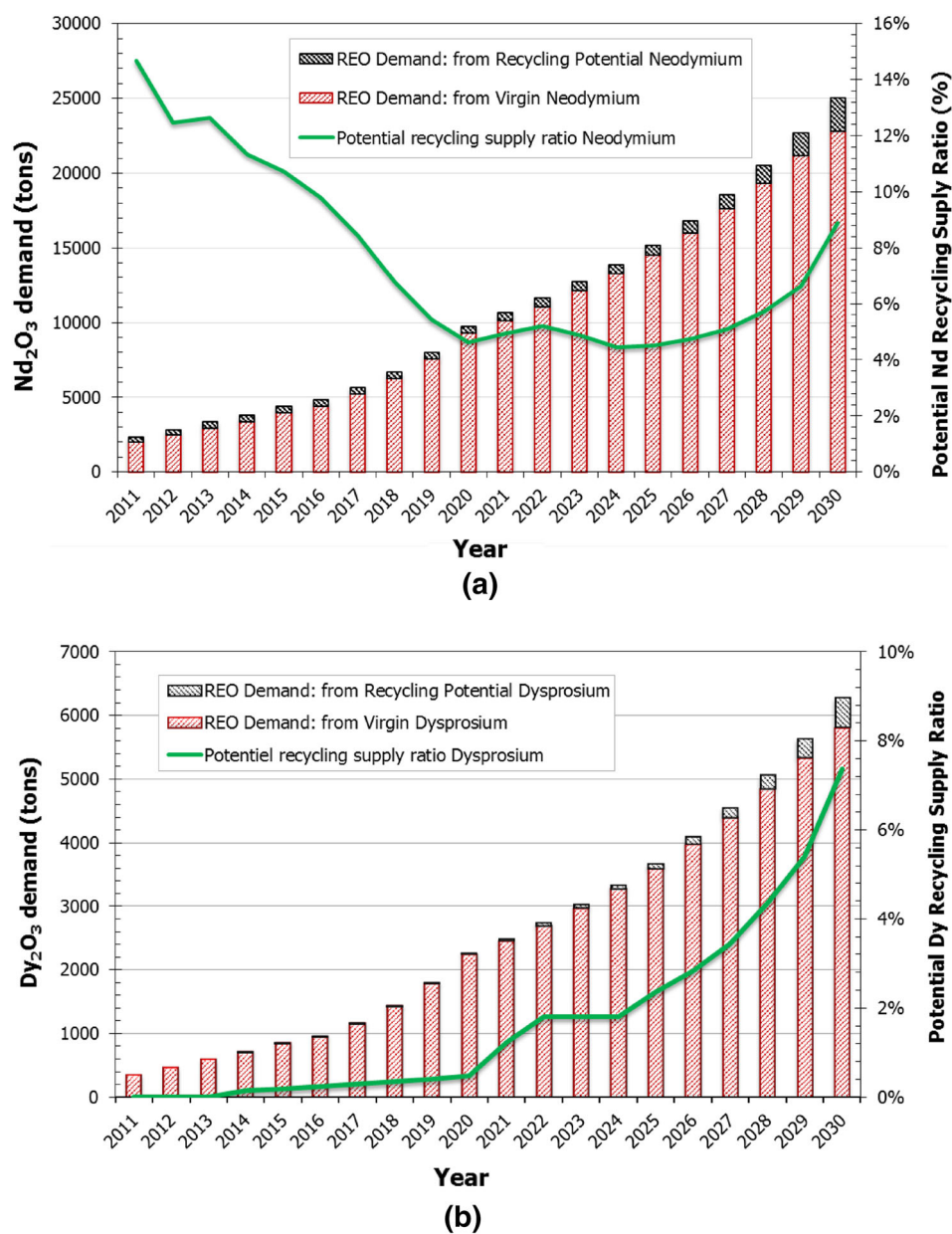
**Table 3** Estimated global in-use stocks for the rare-earth elements in NdFeB permanent magnets in 2007 [49]

| Stocks (kt)          | Nd   | Pr   | Dy   | Tb  | Total | Share (%) |
|----------------------|------|------|------|-----|-------|-----------|
| Computers            | 21.2 | 5.3  | 5.3  | 1.1 | 32.8  | 33.8      |
| Audio systems        | 15.1 | 3.8  | 3.8  | 0.8 | 23.4  | 24.1      |
| Wind turbines        | 10.1 | 2.5  | 2.5  | 0.5 | 15.7  | 16.2      |
| Automobiles          | 9.8  | 2.5  | 2.5  | 0.5 | 15.2  | 15.7      |
| Household appliances | 3.3  | 0.8  | 0.8  | 0.2 | 5.1   | 5.3       |
| MRI                  | 3.0  | 0.8  | 0.8  | 0.2 | 4.7   | 4.8       |
| Total                | 62.6 | 15.7 | 15.7 | 3.1 | 97.0  | ~ 100     |

slag samples indicated that a substantial presence of Nd was detected in the EAF steelmaking slags with an average concentration of 0.032 wt% Nd (320 ppm). The Nd should come from the NdFeB permanent magnets used in small electric motors and acoustic devices in the cars and electrical and electronic appliances.

For the generators in wind turbines, the REE permanent magnets, after reaching their end of first life, can still be manually dismantled and reused or recovered much more easily due to their large size (250–600 kg/MW power). For the conventional automobiles, there are around 40 magnets in small motors and actuators and 20 sensors in a typical

**Fig. 6** Predicted recycling potentials of Nd and Dy from EOL permanent magnets for all the 3 sectors (computer hard disk drives, wind turbines, and automotive industry), adapted from Rademaker et al. [2] (reprinted with permission). **a**  $\text{Nd}_2\text{O}_3$  demand for permanent magnets, **b**  $\text{Dy}_2\text{O}_3$  demand for permanent magnets



automobile, and on average there are about 250 g of NdFeB magnets and 10–20 g of SmCo magnets as stated by Shaw and Constantinides [23]. These small permanent magnets are not pre-dismantled before car shredder, and they are lost to ferrous or nonferrous scrap similar to the magnets in the consumer electronics. The REE permanent magnets in the electric motors EVs and HEVs are relatively larger than in normal cars (on average 1.25 kg, see Table 2) but still difficult to dismantle manually. Depending on the market value, manual dismantling of the REE magnet-containing components in major consumer products could be an option for further mechanical and metallurgical recovery in the future.

### Analysis of REE Magnets in the Shredder Material Streams

#### *Conventional Shredding Operation for ELVs and Household Appliances*

As is discussed earlier, most of the permanent magnets go to shredders together with the bonded EOL products. A recent study by Bandara et al. [46] gives a good overview about the destinations and concentrations of the permanent magnets and the associated REEs. Their study for shredding ELVs and household appliances indicates that the ferrous scrap of the shredder products contains about 130–290 g of Nd/ton, or 130–290 ppm. Figure 7 illustrates the shredder feed compositions and the destination of the REE magnets for a mixed shredding of ELVs, household appliances, and other sources. This concentration is even much lower than the REEs content in some mine tailings which can be up to 1000–1500 ppm. Without pre-concentration and novel technologies, the low REE content in the scrap is currently very difficult to recover in the metal industry. Technologies for pre-shredder separation are needed for feasible metallurgical recovery.

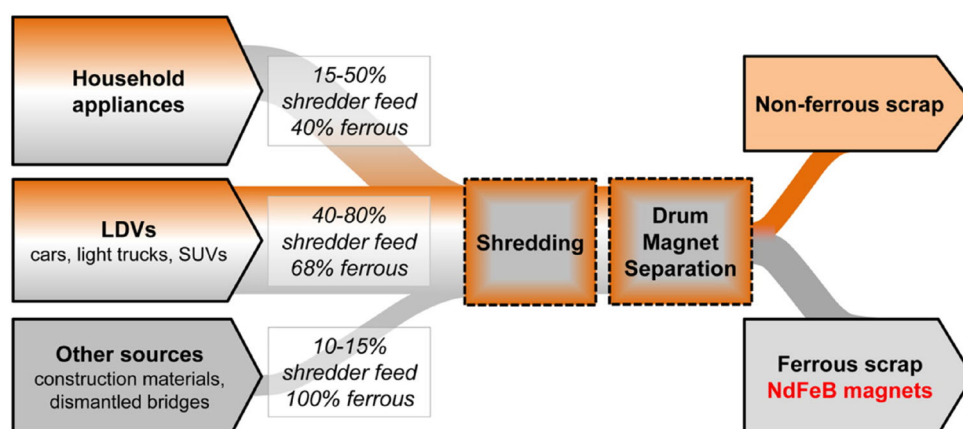
#### *Magnet Scrap From WEEE Shredding*

At the moment, most of the collected WEEEs are shredded without pre-dismantling for the magnets or magnet-containing components such as HDDs from PCs and laptops or speakers from TVs or mobile phones. In principle, the collected WEEEs are directly shredded as a whole after removing batteries or screens, or any toxic/dangerous components and materials. The permanent magnets are normally reported to the ferrous fraction due to its strong magnetism or stick to the shredder systems without separate collection. In this operation mode, the REEs in the permanent magnets have very low concentration and is very difficult to be up-concentrated for economic recovery. Some recycling companies are stockpiling the manually dismantled REE magnet-containing components such as computer HDDs for future REE recovery, since there is no mature or economically feasible REE recover technologies. Shredding of manually dismantled HDDs is normally practiced for data security of data centers and network drives, and the shredded HDDs are sold as ferrous and nonferrous scrap, and thus REEs in the permanent magnet are lost in the subsequent metal recovery in smelters. This will be further discussed in “[Shredding and Physical Upgrading of Pre-dismantled Computer HDDs](#)” section.

Habib et al. [53] studied the flow of resources in electronic waste—with a case of EOL computer hard disk drives. Their results demonstrate the complete loss of REEs in the existing shredding-based WEEE treatment processes. Dismantling and separate processing of NdFeB magnets from their end-use products can be a more preferred option over shredding. However, it remains a technological and logistic challenge for the existing system.

There have been several joint industry–academic efforts in developing separate treatment technologies for computer HDDs and compressors of air conditioners, and this will be described in the “[Separation of Magnets Prior to Shredding](#)” section.

**Fig. 7** Illustration of industrial shredding process for typical EOL products and NdFeB magnets in shredder outputs [46] (reprinted with permission)



of HDDs and Air Conditioning Units” section following this. Furthermore, within an EU FP7 project REEcover (<http://www.reecover.eu>), great efforts are being made to develop economically feasible technologies to recover low concentration REEs in the NdFeB permanent magnets from various standard and specific WEEE shredder products. Industrial operation outputs of various product streams (ferrous, nonferrous, and dust) are sampled and characterized systematically, and further upgrading of REE-rich streams (Nd, Pr, and Dy) originating from NdFeB permanent magnets were intensively investigated using various combination of size reduction and physical separation technologies, including thermal demagnetization, screening, cryo-grinding, and sieving. Quite promising results were obtained and reported in a recent conference paper in 2015 [54]. More detailed publications are expected in the near future, according to the project management.

In order to separate the magnets prior to shredding, several problems have to be overcome. First, the magnets are often held in position with glues and the components held in the electronics with screws. Then the magnets are also often coated with Ni or Ni–Cu–Ni, which has to be removed prior to reprocessing, and finally the magnets have to be demagnetized. Ueberschaar et al. [55] suggested that standardized alloy mixtures for NdFeB magnets, their coatings, and alloy types would help to simplify the recovery of the magnet material for a subsequent use. Several solutions have been proposed, and they are outlined below in [Separation of Magnets Prior to Shredding of HDDs and Air Conditioning Units](#)” section.

### Separation of Magnets Prior to Shredding of HDDs and Air Conditioning Units

#### *Hydrogen decrepitation of Pre-dismantled Computer HDDs*

The University of Birmingham (UK) has developed a hydrogen-processing method to extract sintered NdFeB magnets from computer hard disk drives [13, 41]. During this process, either the voice coil assemblies from the hard disk drive or the cropped corner from the drive is exposed to hydrogen at atmospheric pressure and room temperature. The sintered magnets react with the hydrogen forming  $\text{NdH}_{2.7}$  at the grain boundaries. The grain boundary reaction is exothermic and results in the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  matrix grains forming an interstitial hydride. The overall reaction in hydrogen results in about 5 % volume expansion, and as the magnets are brittle, they break apart resulting in friable hydrogenated powder. The coercivity of the powder becomes so low that the hydrogenated material is essentially soft magnetic and is no longer magnetically attracted to ferrous components in the scrap. This process has been

named Hydrogen Processing of Magnet Scrap (HPMS) [41]. During the HPMS process, when applied to hard disk drives, the nickel coating peels away from the surface of the magnets as thin sheets. After the HDD components are processed in hydrogen, they are rotated in a porous drum in order to liberate the hydrogenated powder from the electronics. At this stage, the powder still contains small nickel particles. The nickel contamination is subsequently removed by mechanically sieving the powder mixture. Figure 8 shows the pilot facilities developed at the University of Birmingham.

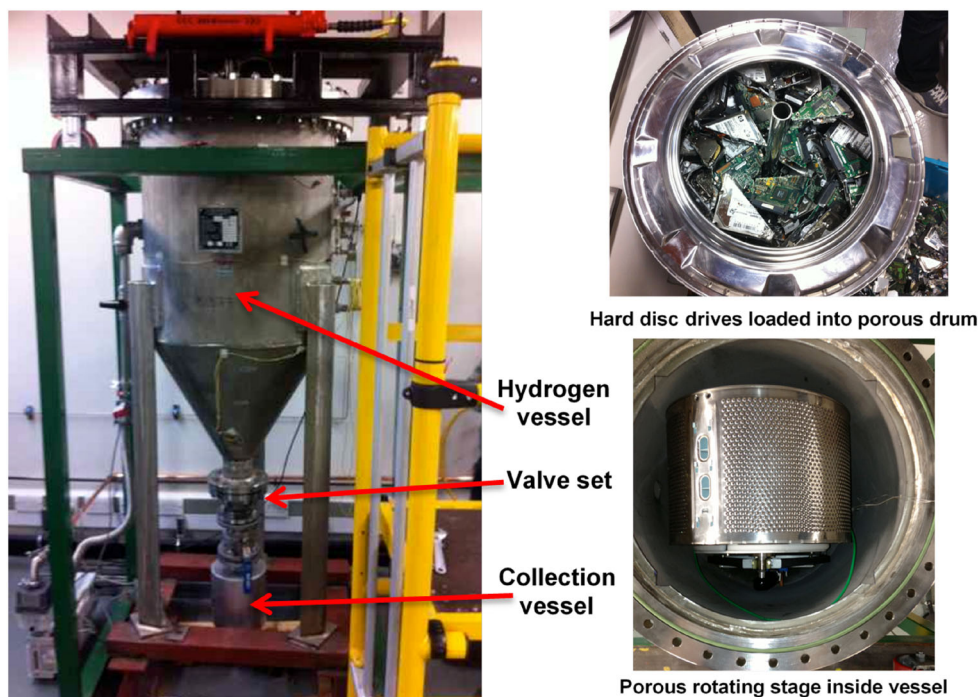
As the hydrogenated powder is much more friable than the nickel flakes, and hence, it mills faster than the nickel, and when passed through a 90- $\mu\text{m}$  sieve, the nickel contamination is reduced from around 2–3 wt% in the starting magnet to below 400 ppm in the final hydrogenated NdFeB powder. This is equivalent to the nickel content of the base alloy, and thus it has entirely been removed. The NdFeB alloy powder which is generated can be used directly to form new NdFeB magnets using a number of routes which are outlined in [“Direct Alloy Recycling Routes”](#) section.

#### *Mechanical Separation of Magnets From HDDs and Air Conditioners*

The Hitachi Group (Japan) developed a mechanical dismantling and separation technique for NdFeB magnets in HDDs and air conditioners, using a rotational drum. Through vibration and impact by tumbling, the screws fixing the HDD casing become loose, and the magnets can be separated out of the casing in about 30 min. The apparatus can process 100 HDDs per batch and 200 HDDs per hour, much faster than manual dismantling at a rate of 12 units per hour [9, 56, 57]. Hitachi and Mitsubishi Materials have also developed similar technology to recover the NdFeB magnets from the compressors in air conditioners. After cutting off the casing, the rotors in the compressor can be removed and treated by either non-thermal demagnetization using resonance damping demagnetization at room temperature [57] or thermal demagnetization at 400–500 °C. Hitachi may use the similar rotating drum for disintegration of HDDs to loosen the magnets from the air conditioners, and Mitsubishi uses other equipment (e.g., drills). Furthermore, Mitsubishi Materials is also trying to separate the NdFeB magnets from direct drive (DD) motors in washing machines through thermal demagnetization and removing the resin bonding [9]. Figure 9 illustrates the commercial process to separate REE magnets from HDDs and other scraps to automatically separate fragments of precious metals or other valuables [57]. According to Baba et al. [57], trials were to be conducted on a commercial scale. However, no



**Fig. 8** A 300-L hydrogen vessel and rotating porous drum for separation of hydrogenated NdFeB powder from HDDs at the University of Birmingham



more recent update about the status of commercialization and operation has been reported to the public.

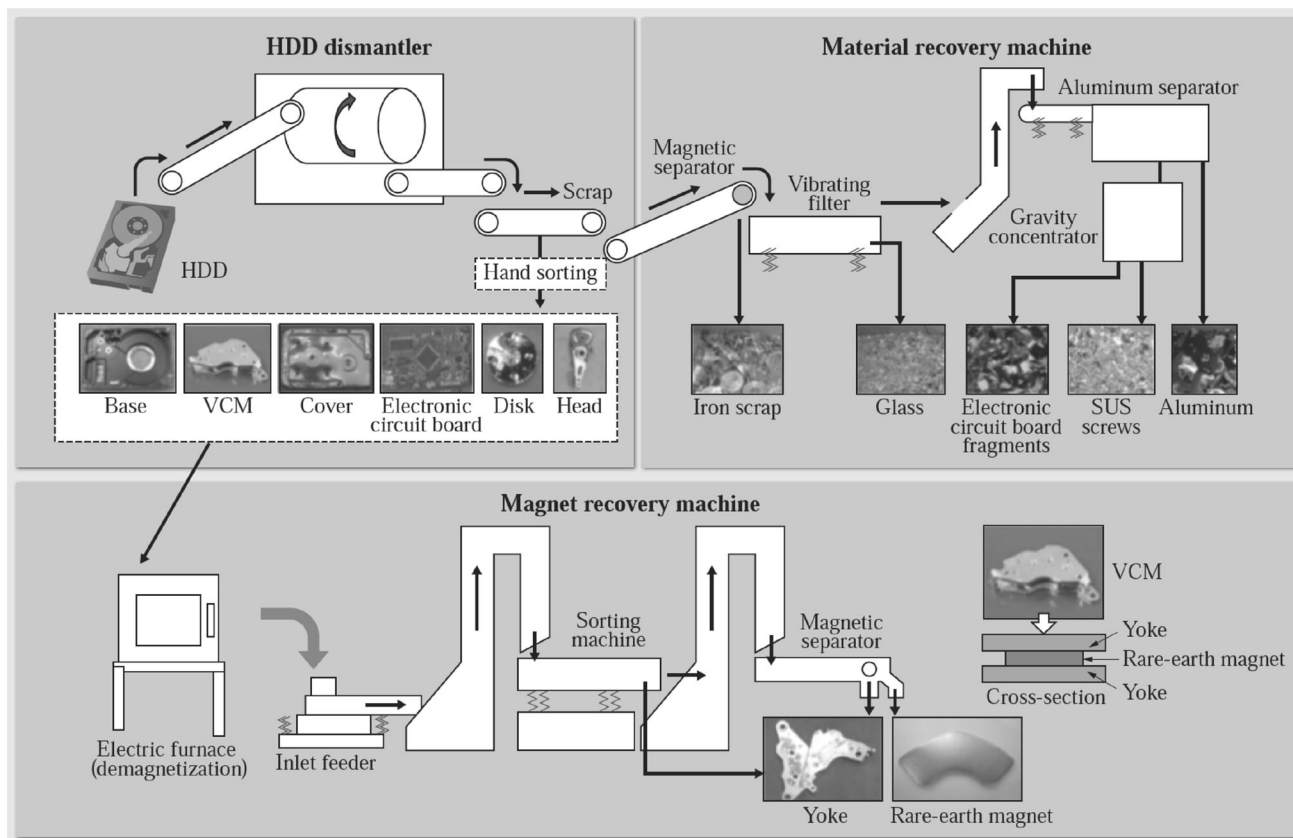
### Shredding and Physical Upgrading of Pre-dismantled Computer HDDs

Pre-dismantling of computer hard disk drives have been reported by some companies mainly for data security reasons, although this is not a standard practice for normal personal computers from offices and homes. The dismantled hard disk drives are shredded, and normally, the ferrous and nonferrous scraps as well as the printed circuit boards (PCBs) are recovered. In most cases, the REE magnets particles after shredding are not recovered although the REE concentration is much higher than those in the shredded ferrous products from the whole EOL product (e.g., computers).

In the Netherlands, Van Gansewinkel Group (VGG) operates a small shredder for HDD shredding (data destruction). TU Delft (The Netherlands) has recently developed a REE-recovery process from the magnet-concentrated HDD shredder residues together with VGG [58, 59]. It was found that a feasible REE-recovery process is dependent on an efficient liberation and separation of the magnets from the other components of the HDDs, thereby minimizing the amount of bulk contaminations. The study indicates that when a coarse shredder was used for this purpose, magnets were poorly liberated from other components. This generated a lot of magnet-bearing pieces after shredding. Although the typical shape of the magnets

or the appearance of scattered dark magnetic particles, enables their selection by visual screening, this collection method is inefficient and not economical, producing large amounts of nonmagnetic materials for further processing. On a contrary, when a fine shredder was applied, magnets were broken into small particles, and the 30-mm steel grate was revealed as an effective tool to retain the magnetic content while still allowing continuous flow of nonmagnetic materials into the outlet stream. Figure 10 shows the grate after HDDs shredding. Even in this case, however, the as-collected grate residue still contains some nonmagnetic metallic pieces. In order to characterize the content of the grate residue, the sample was hand-sorted into nonmagnetic and magnetic materials as illustrated in the upper part of Fig. 9. The total mass ratio of magnet-to-nonmagnet pieces was approximately 2.2. With this approach, approximately 70 % of the magnet particles was collected in the grate for further processing and recovery.

The separately collected magnet residues go through a thermal demagnetization process followed by grinding and screening. After a mild grinding and screening, the magnet concentrate particles of less than 1 mm in diameter represent approximately 63 % of the total collected shredder residue, and a total recovery rate of 95 % has been achieved for all the REEs in the collected residues through physical processing. The concentration of REEs (Nd + Pr) was upgraded from 5 to 7 % in the as-collected fine shredder residues to about 17–20 % after the above-mentioned physical processing. The highly concentrated magnet particles separated from the ductile metal pieces can go



**Fig. 9** Process used to separate and recover REE magnets from HDDs [57] (reprinted with permission)

through either hydrometallurgical extraction or first processed with slag extraction. The REE product of fluoride or oxide can be produced. Figure 11 illustrates the developed processing flowsheet.

### Direct Alloy Recycling Routes

If a clean, nonoxidized form of scrap NdFeB magnets can be separated from the EOL products, in the form of a hydrogenated powder (HPMS), or as a solid magnet, then it is possible to reprocess the material directly from the alloy using a number of routes. It should be noted that these routes are not suitable for shredded material as the contamination levels of this powder will be very high. These direct alloy methods include resintering of the powder, melt spinning, HDDR processing, or recasting back into a master alloy [41]. These routes are used in the primary production of magnets but require some modification in order to handle these secondary materials. For example, a recycled source of sintered NdFeB material will typically contain 2000–5000 ppm oxygen, which is far higher than in a primary cast NdFeB material (typically 300–400 ppm oxygen). Also a recycled source of NdFeB magnets, such as HDD magnets, will contain a range of magnet

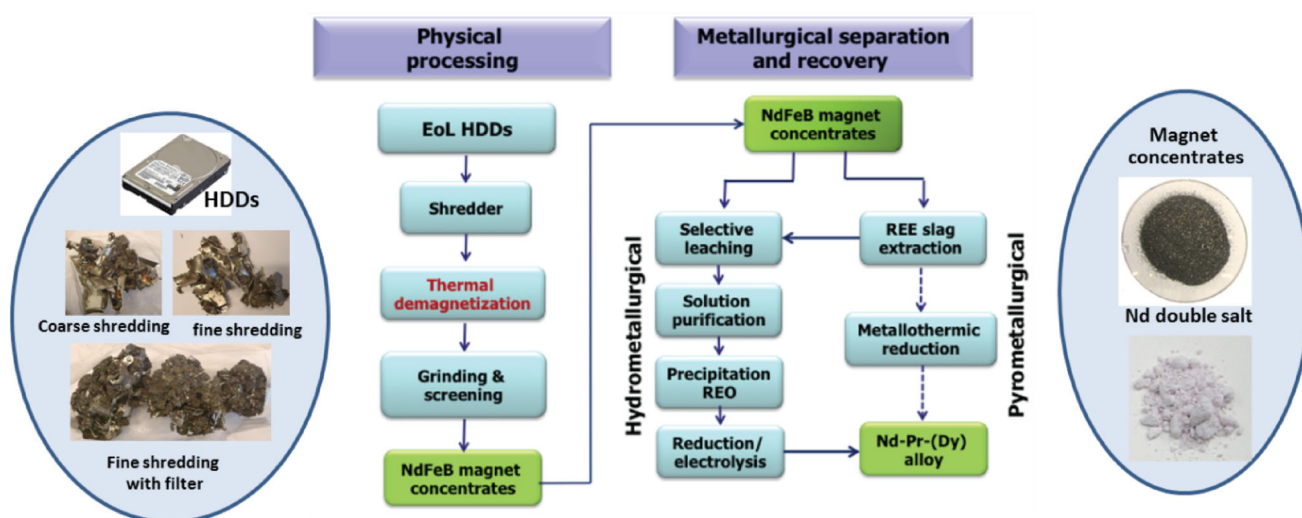
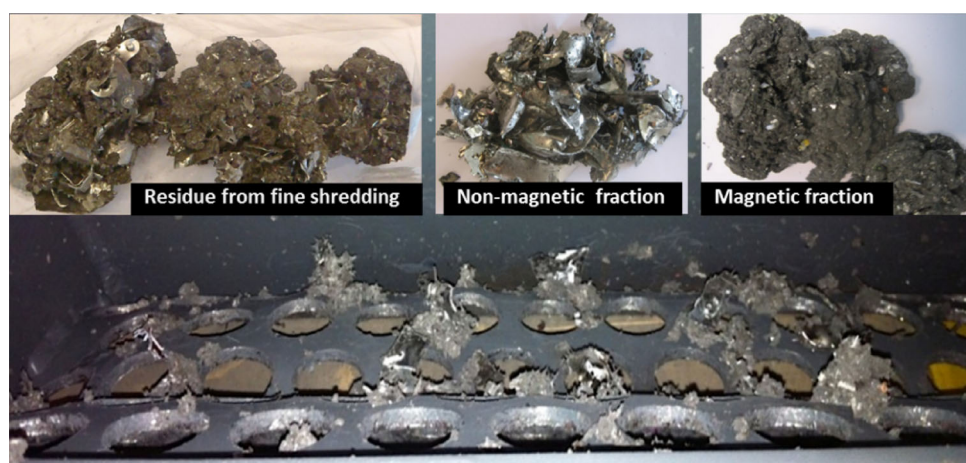
compositions [60], so questions arise about the consistency of magnetic performance, which can be achieved from magnets made from this material. Despite these potential disadvantages, direct alloy production routes will typically have a smaller environmental footprint compared to recycling routes, which rely on stripping of the REEs (see “[Hydrometallurgical Methods](#)” and [REE Separation by Solvent Extraction](#)” sections). As fewer process steps are required to reprocess directly from the alloy then the cost of magnet production will also invariably be smaller.

### Re-sintering of Scrap NdFeB Magnets

Researchers at the University of Birmingham demonstrated that it is possible to produce resintered magnets by processing of NdFeB magnets in hydrogen, blending and milling of the subsequent powder with extra REE material, followed by magnetic alignment of the powder and iso-static pressing to form green compacts. The green compacts were then sintered at  $\sim 1080^\circ\text{C}$  [61, 62]. Further studies by Högberg et al. [63] have shown that it is possible to process solid sintered magnets back into sintered magnets by thermal demagnetization and then milling and sintering, without the use of hydrogen gas. However they



**Fig. 10** Magnetic residues in the grate after fine shredding. Photos from left to right on the top: manually collected residues, separated non-magnetic and magnetic fractions after hand sorting [58, 59]



**Fig. 11** Process flowsheet for the REE recovery from EOL computer hard disk drives, developed at TU Delft

reported irreversible losses using this route due to the heating process [63].

Scrap sintered magnets will typically contain 2000–5000 ppm oxygen and most of the oxygen is entrained in the grain boundary phase [64]. On resintering, the grain boundary phase no longer melts due to the higher melting point of the oxide and therefore full density cannot be achieved in the resintered magnets. Therefore this grain boundary phase needs to be replaced. Zakotnik et al. [61] demonstrated that by powder blending 2 at%  $\text{NdH}_{2.7}$  into the hydrogenated  $\text{NdFeB}$  powder that full density could be achieved in the recycled magnets and further studies by Heraiz et al. [65] have shown that by adding >2 at%  $\text{NdH}_{2.7}$  the coercivity can be higher than the starting magnet. The remanence of the magnets is typically reduced by around 2–10 % due to the extra nonmagnetic volume of the  $\text{NdH}_{2.0.7}$ . In subsequent publications REE metals have been added in the form of alloy powders, which has

allowed for some tailoring of the magnetic properties. For example, by adding an alloy containing Dy to increase coercivity in a similar fashion to its addition in primary  $\text{NdFeB}$  magnets [61]. Powder blending in this way is a common technique in the production of primary  $\text{NdFeB}$  magnets and in a recycled magnet the alloy still supplies extra rare-earth material to aid liquid phase during sintering [66].

Several papers have been published on scaling of the resintering process. For example Zakotnik et al. [67] produced 120 kg of resintered magnets from scrap sintered material which had been pre-separated from the product. However, the variability of remanence, coercivity and maximum energy product are not reported across the entire batch of magnets which were remanufactured. The variability of magnetic performance is as important as the ultimate magnetic performance for the end users of these magnets. Researchers in the EU Framework 7 project

REMANENCE (<http://www.project-remanence.eu/>) have demonstrated that it is possible to extract 5 kg of magnets from ‘real’ scrap hard disk drives provided by Stena Metal in Sweden using the HPMS process, outlined in “[Hydrogen Decrepitation of Pre-dismantled Computer HDDs](#)” section, After jet milling, blending, pressing and then resintering the maximum energy product of the final magnets was shown to vary by only  $\pm 5 \text{ kJ/m}^3$  respectively [68]. This level of variation is similar to that observed in primary magnet production despite the mixed compositional feed which was presented to the process [68].

### Hydrogenation Disproportionation Desorption and Recombination (HDDR) of Scrap-Sintered NdFeB Magnets

It was first demonstrated that the HDDR process could be applied to scrap sintered NdFeB magnets in 2004, by Kim et al. [69] and Zakotnik et al. [70], in order to produce isotropic NdFeB powders suitable for processing into resin-bonded magnets. However further research demonstrated that it was possible to produce anisotropic HDDR powdered material [38, 39]. The HDDR process is essentially the same process as that outlined in “[Properties and Production Methods of Rare-Earth Permanent Magnet \(REPM\)](#)” section, However due to the grain size and compositional variation in the secondary material compared to primary cast material, then the temperature and pressure have to be shifted in order to optimize the magnetic performance of the material [40]. The HDDR powder can then be used to produce anisotropic resin-bonded magnets by cold pressing or injection molding in an aligning field [71, 72]. Li et al. [71] demonstrated that it is possible to produce compression molded resin-bonded magnets with a remanence of 0.9 T, a coercivity of 841.4 kA/m, and a maximum energy product of 111.6 kJ/m<sup>3</sup>. However this work did not address the variability of magnetic performance from ‘real’ mixed scrap feeds. Workers in the EU FP7 project REMANENCE (<http://www.project-remanence.eu/>) have recently demonstrated the use of HDDR recycled powders for the production of injection molded magnets. An amount of 5 kg of hydrogenated powder was produced using the HPMS process. This powder was then HDDR processed at around 900 °C and 1.2 bar H<sub>2</sub> pressure in 400 g batches. The HDDR material was then injection molded with a polyamide binder at Kolektor in Germany in 5 kg batches. It was shown that the coercivity, remanence and maximum energy product only varied by  $\pm 1 \text{ kA/m}$ ,  $\pm 2 \text{ mT}$  and  $0.34 \text{ kJ/m}^3$  respectively across the whole 5 kg, which is again in line with magnets produced from primary cast NdFeB material [73].

### Re-casting and Melt Spinning of Sintered NdFeB Magnet Scrap

Another potential route to reprocess scrap sintered NdFeB magnets is to remelt the scrap sintered magnets by induction heating, in order to convert the material into a master alloy. The master alloys can then be converted back into magnets using conventional primary processes. This route has the advantage that the oxygen concentration can be reduced in the overall material from 2000 to 5000 ppm in the sintered magnet to around 300–700 ppm in the cast alloy [60]. However this process is typically associated with a yield loss of around 20–30 %. This type of reprocessing is a common way of reprocessing sintered production scrap in the magnet industry. The oxygen is removed in the slag phase, however this does also cause significant wear of the crucibles.

It is also possible to convert sintered magnets into melt-spun ribbons, as in “[Properties and Production Methods of Rare-Earth Permanent Magnet \(REPM\)](#)” section, which are suitable to form resin-bonded magnets. [74]. The magnets are induction melted and then the molten material is ejected under an argon pressure onto a rotating copper wheel to produce ribbons of nanocrystalline NdFeB. This route will suffer from similar yield losses compared to the recasting route for master alloys. Itoh et al. [74] demonstrated this route in 2004 and produced magnets with a remanence of about 0.69 T, a coercivity of about 0.70 MA/m, and a maximum energy product of about 71.0 kJ/m<sup>3</sup>.

### Metallurgical Extraction and Separation

Except for the large NdFeB magnets used in wind turbines and electric motors of EVs/HEVs, direct alloy recycling will be difficult and impractical for the rest of applications. Metallurgical extraction, separation and refining are necessary for small REE magnets and the magnets originated from shredded EOL products and even manually dismantled REE magnets from computer HDDs or other consumer products. In the case of computer HDDs, Ueberschaar et al. [55] conducted a detailed study for the recycling and recovery of REEs and other valuable metals from HDDs, and they emphasized that high variations in alloys used for the voice-coil actuator and for the spindle motor magnet conflict with a direct reuse of the magnets or reutilization of the magnet material for resintering processes. Together with alloy composition changes over time due to fluctuating raw material prices and increased demands for physical properties, eventually hydro- or pyrometallurgical recovery processes are needed, after certain levels of dismantling and physical separation and mechanical processing.

There have been many studies for the recovery of Nd and other associated REEs (Pr, Dy, Tb) through different types of metallurgical processes: hydrometallurgical, pyrometallurgical or electrochemical, or combinations of these techniques. However, most of these studies have focused on the highly concentrated and relatively clean waste magnets of manufacturing scrap or production waste: the new solid scrap and cutting sludge “swarf” [75] which accounts for 20–30 % of the starting alloy [9]. The solid scrap is normally remelted (after coating removal) within the magnet manufacturing plant for direct reuse. However, the swarf requires chemical or metallurgical separation and refining mostly through hydrometallurgical leaching route to produce rare-earth fluorides or oxides as raw materials for conventional metal production (molten salt electrolysis or metallothermic reduction). Recently published comprehensive reviews on rare earths’ recycling by Binnemans et al. [8], Tanaka et al. [9], Takeda and Okabe [10], and Firdaus et al. [11] are good sources of reference, covering large variety of metallurgical methods. Binnemans et al. [8] gave a good comparison for different metallurgical methods as is partially listed in Table 4.

For the small magnets used in the EOL products, little recycling efforts are identified and less than 1 % of the EOL REEs (including magnets) has been recycled according to many expert views, mainly due to their small size, tight bonds with the product and the multiple type mixture of different magnets (ferrites, SmCo and NdFeB) [9].

As discussed in “Physical Separation Technologies” section, the magnet or REE concentration is too low for economically feasible metallurgical recovery if the EOL products (automobiles, home electrical appliances and consumer electronics) are directly shredded. Pre-dismantling and up-concentration through physical processing are critical for viable chemical or metallurgical recovery. Recovery of the minor amounts of REEs from the magnet scrap is technically and economically challenging. Allocating the REEs in the magnet scrap into the by-product or waste streams via smelting processes of main metals is a potential possibility, including slags, flue dust, solid leach residues and waste solutions. Therefore, a total materials recovery from EOL products (containing REE magnets) would be the direction for future research and development, in parallel with magnet pre-dismantling approach.

### **Hydrometallurgical Methods**

The key steps for hydrometallurgical route are leaching of the magnets or magnet scrap, followed by separation of individual REE species (Nd, Pr, Dy) using solvent extraction, ion exchange, or ionic liquid techniques. Afterward, either selective precipitation of the mixed REEs

from co-dissolved non-REE species, or precipitation of individual REE salts or compounds are required, and finally conversion to REE fluorides or oxides could be made. The challenges for the hydrometallurgical process to recover the REEs in the EOL permanent magnets, in particular the REE dilute waste streams, are the selective dissolution, up-concentration and separation of REE species from the major species, and total recovery of all major metals at the same time. Therefore, new separation technologies using novel solvent extractants, ionic liquid, ion exchange resin or the alike, are needed to extract REE species from low concentration leaching solutions.

### *Leaching of NdFeB Magnet Scrap*

Leaching is always the first step to dissolve the REEs in the magnet or magnet scrap, preferably prior to other metals in the magnet and magnet scrap. Depending on the complexity level, different dissolution methods could be used. Dissolution of magnet scrap can be performed in three different ways: (1) complete dissolution of the NdFeB magnet (with or without a prior roasting step), (2) roasting followed by selective leaching of the REEs, and (3) selective conversion of REEs in solid magnet or magnet scrap directly to a new solid phase. Alternatively, transformation of REEs in the magnet scrap into REE compound precipitates could be realized, based on solubility of REE salt at different temperatures or under hydrothermal conditions.

*Complete leaching (with/without roasting)*  $\text{H}_2\text{SO}_4$  leaching at room temperature has been developed by Layman and Palmer in 1993 [17] of US Department of the Interior Bureau of Mines in 1993 for relatively clean production scrap. All components in NdFeB magnets will be dissolved. Nd can be precipitated as double sulfate salt which could be easily converted to  $\text{NdF}_3$  or  $\text{Nd}_2\text{O}_3$ . However, large amount of Fe dissolved in the solution needs to be properly disposed (jarosite precipitation). Recently Abrahams et al. [59] successfully applied a similar  $\text{H}_2\text{SO}_4$  leaching for the treatment of complex EOL NdFeB magnet scrap from an HDD shredder. Very recently Bandara et al. [76] have demonstrated a HCl leaching process to selectively dissolve NdFeB magnet from shredded electric motors—a mixture of REE magnet, (stainless) steel and copper. The thermally demagnetized magnet particles were completely dissolved in 4 M HCl solution for 24 h at room temperature, while the (stainless) steel and copper particles remained completely unreacted. The leach solution was subsequently treated with oxalic acid to precipitate REEs (Nd, Pr, and Dy) as REE oxalate, and the effluent containing Fe and other dissolved transition metals was suggested conceptually to go through a pyrohydrolysis process to generate iron oxide and HCl

**Table 4** Comparison of different metallurgical methods for REE recovery from NdFeB magnet scrap, adapted from Binnemans et al. [8]

| Methods                                       | Advantages  | Disadvantages   |
|---|---|---|
| Hydrometallurgical                            | Generally applicable to all types of magnet compositions<br>Applicable to nonoxidized and oxidized alloys<br>Same processing steps as those for extraction of rare earths from primary ores   | Many process steps required before obtaining new magnets<br>Consumption of large amount of chemicals<br>Generation of large amount of waste water and effluents   |
| Pyrometallurgical:<br>Liquid-phase processing | Generally applicable to all types of magnet compositions<br>No generation of waste water and effluents<br>Fewer processing steps versus hydrometallurgical<br>Direct melting for obtaining master alloys<br>Liquid metal extraction allows for obtaining REEs in metallic state | High energy input required<br>Direct smelting and liquid metal extraction cannot be applied to oxidized magnets<br>Electroslag refining and the glass slag method generate large amounts of solid waste |
| Pyrometallurgical: gas-phase extraction       | Generally applicable to all types of magnet compositions<br>Applicable to nonoxidized and oxidized alloys<br>No generation of waste water and effluents   | Consumption of large amount of chlorine gas<br>Aluminum chloride is very corrosive  |

back to leaching. A total of 82 % REE recovery has been reached from the total processing from shredding to oxalic precipitation with a product purity of 99.8 % of REE oxalate.

Yoon et al. [77] used an oxidative roasting at 500 °C for sintered scrap and at 700 °C for bonded scrap followed by  $\text{H}_2\text{SO}_4$  (2 mol/L) leaching at 50 °C for 2 h and double salt precipitation. Dissolution of  $\text{Nd}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  seem to be much faster than direct leaching of the magnet, but there is no selectivity of leaching  $\text{Nd}_2\text{O}_3$  over  $\text{Fe}_2\text{O}_3$ . The achieved recovery of Nd yield was over 99.9 %, but about 95 % of iron was also dissolved similar to Layman and Palmer [17]. HCl could also be used for complete magnet dissolution [78]. Subsequently, REEs can be precipitated from leach solutions by adding oxalic acid or hydrogen fluoride to form oxides or fluorides.

**Selective leaching (with roasting)** Selective Nd leaching could be obtained by first roasting of the magnet in air at 700 °C followed by  $\text{H}_2\text{SO}_4$  (4 mol/L) of  $\text{Nd}_2\text{O}_3$  by controlling pH at 70 °C for 3 h with pulp density of 100 kg/m<sup>3</sup> [79]. However, it reached only 70 % Nd leaching recovery. Formation of NdFeO<sub>3</sub> at higher roasting temperatures may hinder the dissolution of Nd. Selective leaching after oxidative roasting at 900 °C for 6 h was also reported by subsequent dissolution with HCl (0.02 mol/L) at 180 °C for 2 h [80, 81]. More than 99 % REE and less than 5 % Fe recovery have been reached.

**Hydrothermal conversion** Itakura et al. [82] investigated the hydrothermal treatment of Nd–Fe–B sintered magnet using a mixture of hydrochloric and oxalic acids. Hydrothermal treatment is an in situ conversion method to change one mineral to another under hydrothermal

conditions (high temperature and high pressure). They used a mixture of HCl (3 mol/L) and oxalic acid (0.2 mol/L) to recover Nd as oxalate. A Nd conversion of over 99 % was achieved at 110 °C for 6 h in the magnet recovered as solid precipitate of  $\text{Nd}_2(\text{C}_2\text{O}_4)_3$  with the purity of 99.8 %.

**Electrochemical dissolution** At TU Delft, a new method using anodic dissolution of NdFeB permanent magnet scrap is under investigation to dissolve REE species electrochemically [83], separating non-REEs in the scrap into anode slimes or precipitated onto the cathode, through accurate control of electrode and cell potentials, current density on the electrodes, and electrolyte chemistry. It is proved technical feasible to dissolve the REEs from the magnet anode, and simultaneously dissolve ferrous metals and reprecipitate them as oxides/hydroxides in the solution or as metals on the cathode.

Leaching methods offer the flexibility of magnet and magnet scrap dissolution. However, selective leaching of REEs in the magnet scrap is challenging due to the presence of large amount of Fe in the magnet alloy and in the scrap. Quite often a high temperature treatment is needed to increase the selectivity of REEs over Fe and other transition metals. For more detailed description and more leaching methods applied to NdFeB magnet scrap, please see the recent reviews by Binnemans et al. [8] and Tanaka et al. [9].

#### REE Separation Through Solvent Extraction

Solvent-extraction separation of ions from an aqueous solution is based on the formation of complexes between ligand molecules soluble in an organic phase and ions



present in an aqueous phase and the transfer of these complexes to the immiscible organic phase. The distribution ratio ( $D$ ) represents the distribution of the ion of interest between the two immiscible phases. It is defined as the ratio of the total concentration of the ion/element  $A$  in the organic phase and the total concentration of  $A$  in the aqueous phase ( $D_A = [A]_{\text{org}}/[A]_{\text{aq}}$ ). The separation factor (SF), defined as  $SF_{A/B} = D_A/D_B$ , is used to show the degree of separation between two solutes  $A$  and  $B$  in an extraction system. It is important to understand that the distribution ratio of a metal depends both on the chemical equilibria between the different forms of the metal in both phases and on the Gibbs free energy change which occurs when a metal containing species is transferred from one phase to the other. The efficiency of solvent-extraction processes depends on the chemical properties of the ions to be separated, the properties of the ligands and the solvent that they are present in (often called the diluent), the pH of the aqueous phase, the temperature, and other parameters, such as the efficiency of mixing between the aqueous phase and the organic phase and the time of contact between these phases.

The body of literature on specific solvent-extraction processes and extractant systems for separation of REEs is very large. Separation of lanthanides from actinides has been of major interest in the nuclear industry where it is necessary to separate the lanthanides from the actinides. This has inspired to a very large research effort since the 1950s. One of the difficulties in separating the REEs, or lanthanides, by methods like solvent extraction is caused by the fact that they are closely related to each other based on their chemical properties [84]. They are located in the f-block in the periodic table and due to the consecutive filling of 4f-orbitals going from Ce toward Lu, while having the same number of 6 s electrons, the properties of these elements are very similar even though the shielding of the increased positive charge from the nucleus by the electrons create differences in atom radii (“the lanthanide contraction”). The oxidation state +3 is the most common one, both in solid compounds and in solutions. One result of the similarity is that the lanthanides occur together in ores. The similarities in thermodynamic properties influences the formation of complexes in aqueous and organic phases and thus the efficiency of solvent-extraction separation systems. Small differences in the shielding of the charge of the nucleus by 4f electrons between lanthanide ions have to be used to create systems for separation of these ions from each other or in groups [85, 86]. In the production of pure REEs from virgin ores containing mixtures of them with additional elements this generally means that the solvent-extraction separation process must include many mixer settler steps in order to give a sufficient purity of the products. Recycling of REEs from spent

products, such as permanent magnets, by leaching and solvent extraction presents an interesting opportunity since the REEs present in the system have been used as a group and can be recovered and reused as a similar group. However, if the recycling is supposed to produce pure REEs full separation needs to be done.

In industrial solvent-extraction separation of REEs the most common ligands are organophosphorous compounds, such as the di(2-ethylhexyl)-phosphoric acid called D2EHPA [87]. Typical separation factors between adjacent REEs using DEHPA is around 2.5 [88]. Separations of REEs from solutions based on different acids by DEHPA have been investigated by several groups showing that aqueous phases based on hydrochloric and sulfuric acids give better results than those based on nitric acid [89–91]. Other organophosphorous ligands that have attracted a lot of interest for separation of lanthanides are saponified 2-ethyl-hexyl phosphonic mono-2-ethyl-hexyl ester (EHEPA or PC88A) and bis(2,4,4-trimethylpentyl) phosphinic acid (Cyanex 272) [92]. EHEPA was recently compared with D2EHPA for separation of Nd, Dy, and Y from hydrochloric acid solution by Mohammadi and co-workers [9]. The results showed that both ligands separately and in mixtures extracted the metals in the order of preference  $Y(\text{III}) > Dy(\text{III}) > Nd(\text{III})$ . Both ligands are acidic, which means that  $H^+$ -ions are released in the formation of REE-ligand complexes. This introduces the pH of the aqueous phase as a factor that needs to be controlled when controlling a process. Depending on the concentration of REE ions in the solution pure D2EHPA or a mixture of D2EHPA and EHEPA gave the best extraction efficiencies. Different complex stoichiometry dominates depending on the relation between REE and ligand concentrations in the respective phases. The best separation between Nd and Dy was achieved using pure D2EHPA.

A rather detailed review about alternative solvent-extraction routes for separation of REEs from permanent magnets has recently been published by Yoon and co-workers [92]. The group of extractants most used in industry are the organophosphorous compounds di-(2-ethylhexyl)phosphoric acid (D2EHPA), saponified 2-ethyl-hexyl phosphonic mono-2-ethyl-hexyl ester (PC88A) and bis (2,4,4-trimethylpentyl) phosphinic acid (Cyanex 272). It has, for example, been reported that the extraction capability for REEs by organophosphorus extractants decreases in the order  $D2EHPA > PC88A$  (EHEPA)  $>$  Cyanex 272. However, the separation factors between adjacent REEs have been shown to vary according to  $Cyanex\ 272 > PC88A > DEHPA$  [93]. Although Cyanex 272 gives the highest separation factor between Pr and Nd of the three extractants compared by Banda and co-workers, the total extraction percentage of REEs by Cyanex 272 was low. Therefore, in order to obtain high separation



factor together with high extraction efficiency, solvent-extraction experiments separating Nd and Pr from chloride solution containing both metals in a mixture have been performed by Liu and co-workers using mixtures of Cyanex 272 and tertiary amines [94]. A combination of Cyanex 272 and Alamine 336 gave the best synergy. Although the separation factors of the two metals were not increased, the extraction of both metals was enhanced. Organophosphorous extractants and amines have been compared as single extractants by for example Abreu and Morais [95] showing that the organophosphorous compounds were the most efficient. The conclusion from the review of Yoon and co-workers [92] was that DEHPA and PC88A are the most effective and promising extractants for the separations needed for recycling of magnet REEs with PC88A seen as the most promising. In both cases, kerosene can preferably be used as diluent. A hydrometallurgical process for the recycling of NdFeB magnet waste with focus on the separation of Tb and Dy from Pr and Nd by solvent extraction with PC-88A was recently presented by a group in Germany [96]. Results from tests in a laboratory size mixer-settler plant showed that twelve stages are necessary for nearly complete separation (4 N purity, rare-earth element basis). The authors state that although further optimization can be done, the process already shows significant advantages over the primary production route used in industry. The fact that the recent laboratory investigation of these ligands by Mohammadi and co-workers came to the conclusion that DEHPA is superior to PC88A [89] shows that these systems are complex and that many factors influence the results.

One example of an amide compound that has been shown to be valuable as a ligand or co-extractant in solvent-extraction systems for REE separation is *N,N,N',N'*-tetraoctyl-diglycolamide, also called TODGA. TODGA is a solvating extractant, which creates strong tridentate complexes with metal ions and has shown particularly good extraction properties for REEs ions in terms of selectivity in comparison to other ions in the aqueous solution [97]. This extractant has shown good stability at room temperature and miscibility with the commonly used diluents. A disadvantage for large-scale industrial use is currently the price of this reagent.

The recycling of metals from spent products presents new challenges in the form of dilute flows of REEs mixed with other elements that are not present in the ores used for primary production. Therefore, new separation technologies such as novel extractants for solvent-extraction separations are needed to extract REE species from low concentration leaching solutions and to produce REEs in pure compounds. As stated by Tekda and Okabe in a review on the current status of resource and recycling technologies for rare earths from 2014: “It is (also)

important to establish a recycling-based society from the perspective of the conservation of finite and valuable mineral resources and the reduction of the environmental load associated with mining and smelting” [10]. In that context, the chemistry of the ligands used in the solvent-extraction processes for REE production and recycling needs to be taken into account. If possible, it would be beneficial to use ligands based on combustible (leaving no ash products) ligands such as those designed according to the CHON-principle (ligands containing only the elements C, H, O and N) instead of phosphorous or sulfur containing ones. For some time a considerable effort has been put into design and synthesis of efficient reagents complying with the ‘CHON’ principle for the solvent extraction of trivalent f-block elements [98]. An important advance has been the development of malonamides as extractants for lanthanides and actinides. The comparatively easy synthesis of the malonamides, their selectivity for trivalent f-block metals and the fact that they are built according to the ‘CHON’ principle has contributed to, for example, the DIAMEX process [99]. One disadvantage of the malonamides, however, is that the distribution coefficients (ratios) of f-block metal ions are not very high [100]. By increasing the distribution ratio during the extraction part of a purification process, the number of stages of countercurrent extraction required can be reduced thus reducing the size and cost of the plant. Novel extractants for magnet REE solvent-extraction separation are presently being developed at Chalmers University in Gothenburg (Sweden) as a part of the efforts toward better lanthanide recycling options. Three polyamides (multicoordinate, neutral ligands) bearing tetrabutylmalonamide functional groups have been synthesized and evaluated for use as reagents for the solvent extraction of rare-earth elements from nitrate media. These extractants are based on the CHON chemistry, avoiding phosphorous and other ash forming elements which makes it easier to combust the extractant and the diluent once it can no longer be recycled into the process. The results showed good effects of structural modifications of tetrabutylmalonamide on the extraction of  $^{152}\text{Eu}$ , which was used as a model for lanthanides due to that it allows for precise radio-analytical (radiotracer) determination. The best distribution ratios for Eu were obtained with the tetraamide 2,2'-(1,2-phenylenebis(methylene))bis(*N,N,N',N'*-tetrabutylmalonamide). As for other malonamides a diluent with quite high polarity is favorable for solvent extraction of rare-earth metals [101]. It is important to note that oxidation in supercritical water has been shown to be able to process tributyl phosphate, so in the event that a CHON-extraction system for the recycling of the lanthanides cannot be found then an industrial scale recycling process will still be able to operate without recourse to the incineration of phosphorus rich organic wastes. However the

absence of phosphorus and sulfur in the organic waste produced by a solvent-extraction plant would increase the value of such a waste as a fuel.

In some cases, it may be beneficial to have a stationary phase containing the complexation ligands. One example is presented in the work by Kim et al. [102], which reported in 2015, an application of *membrane-assisted solvent extraction* (MSX) to recover Nd, Pr, and Dy from commercial NdFeB magnets and industrial scrap magnets. A hollow hydrophobic polypropylene fiber membrane module system was used to extract REEs in a single step with the feed and strip solutions circulating continuously through the MSX system. The pores of hollow fibers in the membrane modules were filled with organic phase consisting of the TODGA, Isopar L (An aliphatic diluent), and TBP in the ratio of 3:4:3, respectively. The shell side of hollow fibers was supplied with the aqueous strip solution (0.2 M HNO<sub>3</sub>), and the lumen side of the hollow fibers was fed with the REE-containing solution (1000–2000 ppm) in 6 M HNO<sub>3</sub>. They found that a multimembrane module configuration with REEs dissolved in HNO<sub>3</sub> solutions showed high selectivity for REE extraction with no co-extraction of non-REEs, while the use of HCl solution resulted in co-extraction of non-REEs. REE oxides in highly pure form were produced from the strip solutions. It seems that the application of membrane-assisted solvent extraction to the REE recovery from the scrap magnets would result in a more environmentally friendly and cost-effective process compared with the conventional routes such as precipitation and solvent extraction.

### REE Separation by Ionic Liquids

The replacement of the organic phase in solvent-extraction processes is a novel approach in hydrometallurgy. Ionic liquids are solvents that consist entirely of ions. They are nonvolatile and nonflammable so that they can be safer alternatives for molecular organic solvents in extraction processes, including processes for the separation of rare earths [103–107]. The very high concentration of extractants in the case of using undiluted ionic liquids that act as basic extractants (anion exchangers) can lead to very high loadings of metals in the ionic liquid phase and thus to process intensification (less solvent has to be used). The mechanism of solvent extraction with ionic liquids can be different from solvent extraction with molecular solvents, and this offers an opportunity for the development of new more selective separation processes. There are several challenges to deal with when designing ionic-liquid-based solvent-extraction processes: (1) the high viscosity of the ionic liquids phases which hampers mass transport; (2) the tendency of the ionic liquid to extract via an ion exchange mechanism, resulting in losses of the ionic liquid

components to the aqueous phase; (3) the hydrolytic instability of some fluorinated anions such as hexafluorophosphate; (4) the recyclability of ionic liquids; (5) (cyto)toxicity issues; (6) the high price of most types of ionic liquids. Examples of REE separation systems relevant to magnets recycling making use of combinations of ionic liquids and extractants are (1) 1-methyl-3-octyl imidazolium bis(trifluoromethylsulfonyl)imide [C<sub>8</sub>mim][Tf<sub>2</sub>N] in combination with *N,N*-dioctyldiglycolamid acid (DODGAA) [105] and (2) 1-alkyl-3-methylimidazolium hexafluorophosphate [C<sub>n</sub>mim][PF<sub>6</sub>] (*n* = 2,4) in combination with D2EHPA, which is one of the most frequently used extractants for conventional solvent extraction of REEs [108]. DODGAA is a modification of the extractant TODGA which has proven to have a high affinity to lanthanides. In systems with the ionizable DODGAA the REE ions are extracted as 1:3 complexes and the metal ions can be completely stripped by acid solution. This is a significant improvement since the stripping has been a troublesome issue for many systems based on ionic liquids, such as TODGA [109–111]. Kikuchi et al. [112] investigated the extraction of the REE ions Pr(III), Nd(III) and Dy(III) with tributylphosphate (TBP) in the ionic liquid tricaprylmethylammonium nitrate [A336][NO<sub>3</sub>].

During the recent years, the Binnemans group at KU Leuven (Belgium) has made major research efforts to develop sustainable ionic liquid processes for the recovery of REEs from end-of-life NdFeB magnets. The focus of this work is on the use of hydrophobic ionic liquids with nonfluorinated anions (chloride or nitrate). By increasing the alkyl chain lengths on the cation, the ionic liquids can be made hydrophobic (immiscible with water) without the need of using hydrophobic fluorinated anions. The ionic liquids are typically quaternary ammonium or phosphonium ionic liquids such as Aliquat<sup>®</sup> 336 or trihexyl(tetradecyl)phosphonium chloride (Cyphos<sup>®</sup> IL 101). In case the metal ions are extracted via an anion exchange mechanism, the loss of chloride or nitrate ions from the ionic liquid phase to the organic phase is not a major issue. The ionic liquids are used in undiluted form, i.e., without addition of molecular diluents, to take full advantage of the ionic liquid properties. The selectivity of the extraction process can be tuned by the choice of the ionic liquid anion. For instance, transition metal ions are extracted from a chloride aqueous solution to a chloride ionic liquid phase leaving the rare-earth ions behind in the aqueous solution [113], while the reverse situation holds for extraction from a nitrate aqueous phase to a nitrate ionic liquid [114]. After removal of the transition metals by extraction with a chloride ionic liquid, the REEs can be recovered from the raffinate by precipitation with oxalic acid, followed by calcination of the oxalates to oxides [115]. An efficient process was developed to extract Nd and Dy from a nitrate

solution, leaving Co behind, followed by separation of Nd/Dy by selective stripping with EDTA [116]. Dupont and Binnemans [117] developed a new recycling process for (microwave) roasted NdFeB magnets, based on the carboxyl-functionalized ionic liquid betainium bis(trifluoromethylsulfonyl)imide, [Hbet][Tf<sub>2</sub>N]. This ionic liquid shows a temperature-dependent miscibility with water (=thermomorphic behavior): the [Hbet][Tf<sub>2</sub>N]–H<sub>2</sub>O system is biphasic below 56 °C, but it forms one single-phase below this temperature. Using the thermomorphic properties of the [Hbet][Tf<sub>2</sub>N]–H<sub>2</sub>O system, a combined leaching/extraction step was designed. The change from a homogeneous system during leaching (80 °C) to a biphasic system at room temperature causes the dissolved metal ions to distribute themselves among the two phases: Fe is reported to be present in the ionic liquid phase, while Nd, Dy, and Co remain in the aqueous phase. By selective stripping and precipitation steps, the REEs could be recovered as a mixture of Nd<sub>2</sub>O<sub>3</sub>/Dy<sub>2</sub>O<sub>3</sub> (to be separated in a separate process), cobalt is recovered as CoO, and the ionic liquid [Hbet][Tf<sub>2</sub>N] could be regenerated for reuse.

### Pyrometallurgical Methods

The main purpose of high-temperature processing of NdFeB magnet scrap is to selectively convert REEs in the magnet into another phase which separates the main non-REE components. The separated REEs in the other phase is more concentrated and can be used for RE metal production through molten salt electrolysis or metallothermic reduction. The identified pyrometallurgical extraction methods can be divided into the following groups: (1) roasting, (2) liquid metal extraction, (3) molten salt extraction, (4) molten slag extraction, and (5) electrochemical processing. As can be seen from the discussions below, most of the pyrometallurgical methods are applicable to only highly concentrated magnet scrap (production waste: off-specs and swarf), and adaptation is required to treat the dilute EOL magnet scrap. Slag extraction is a promising method to separate and up-concentrate the REEs in the low concentration magnet-bearing scrap such as shredder residues, however, subsequent hydrometallurgical process steps such as REE leaching are needed. More detailed description on high-temperature recovery from NdFeB magnet wastes can be found in a few recent reviews [8–11]. Firdaus et al. [11] emphasized that high-temperature processing is one alternative that can be used to avoid large consumption of water and production of hazardous waste in the recovery process. A number of techniques are available for recovering REEs from magnet waste by high-temperature processing, but all are still at the research stage, and currently none has been applied on a commercial scale in industry. It was also indicated that combinations of

methods may be required to completely recover REEs from magnet waste.

### Roasting

The aim of roasting is to change the form or state of REEs in the NdFeB magnets at high temperatures, so that the roast product could be more selectively dissolved hydrometallurgically for REEs, leaving Fe and other constituents in the magnet and the scrap stream in the solid residue. After roasting, a more effective separation could be reached in the subsequent hydrometallurgical processing.

**Chlorination roasting** For selective chlorination of REEs in the magnet, various chlorination agents have been investigated mainly by the Japanese researchers: using metal halides such as MgCl<sub>2</sub> to convert the REEs into molten NdCl<sub>3</sub> and DyCl<sub>3</sub> at 1000 °C [118], using FeCl<sub>2</sub> at 800 °C [14], using NH<sub>4</sub>Cl at much lower temperature of 300 °C [15], or using Cl<sub>2</sub> gas and AlCl<sub>3</sub> (the so-called chemical vapor transport process) [119]. In most of these cases, the RECl<sub>3</sub> (NdCl<sub>3</sub> and DyCl<sub>3</sub>) were distilled at elevated temperatures to separate from the Fe-based residue. For all chlorination approaches, selectivity and reaction rates are key issues, in particular when treating highly contaminated and partially oxidized magnet scrap.

**Sulfation roasting** During sulfation roasting, the metallic elements are sulfated to form water-soluble sulfates, preferably for REE only, so that the non-REEs such as iron will be restricted at the solid state in oxide form. There are two steps for this process [120]: in the first step, the whole magnetic material is converted into sulfates by digestion in concentrated H<sub>2</sub>SO<sub>4</sub> at room temperature; in the second step, the resulting mixture is roasted at a temperature at which the REE sulfates are still thermally stable, but the iron sulfates decompose into iron oxide insoluble in water.

Taking advantage of the difference in temperature stability of REE sulfates and iron sulfates, the process of sulfation roasting, followed by water leaching allows the separation of more than 95 % of REE in the leaching solution with no iron present. Iron remains in the leaching residue forming a marketable hematite-dominated by-product. This hematite (Fe<sub>2</sub>O<sub>3</sub>) could be used as a red pigment. Impurities such as Co and Ni were also reported to be present in the residue, thus enabling the production of a leaching liquid with at least 98 % REE purity. SO<sub>3</sub> gas released by thermal decomposition of the iron sulfates can be recovered and used for production of H<sub>2</sub>SO<sub>4</sub>. In this way, a cyclic process can be developed, minimizing emission of SO<sub>3</sub>.

**Oxidation roasting** The objective of oxidation roasting is to convert the REE magnet into oxides for all metallic elements, and the subsequent leaching process of REEs

becomes easier or faster, or more selective. The oxidation roasting—REE leaching has been studied by many researchers [77–81], and it has been discussed in the previous section for hydrometallurgical leaching process. It is important to combine the leaching selectivity and roasting conditions.

#### *Selective Extraction of REEs Through Melt Processing*

In this type of method, magnet scrap is treated with either a molten metal, a molten salt, or a molten oxide slag. The REEs in the magnet will be selectively converted or dissolved to another phase, leaving the iron, boron and other contaminating metals as unreacted solid or as a new molten ferrous alloy. This leads to the separation of REEs from the rest of the magnet and magnet scrap.

**Liquid metal extraction** The Ames Laboratory (USA) has developed a liquid metal-extraction process to selectively recover REEs from NdFeB and other REE-bearing alloys using Mg as extractant [121], and later the process was studied further by other researchers, [16, 122, 123]. In this process, the molten Mg and potentially other alkaline earth metals (Ca or Ba) claimed in Ref. [121] can dissolve selectively Nd and other REEs such as Pr and Dy at about 1000 °C (above the melting point of the extractant metal), leaving Fe and B or other transition metal unreacted in solid state. The formed molten Mg–REE alloy could be separated from the iron and other transition metals (in solid state). The Mg–REE separation can be realized through distillation to evaporate Mg metal due to its high volatility. However, this process is very slow (24–72 h), and it cannot be applied to (partly) oxidized NdFeB scrap. Alternatively, selective extraction of Nd was also reported successfully using silver [124] and copper [125]. Using Ag as the extractant, a Nd–Ag alloy containing 45–50 at % Nd was obtained in the temperature range of 1000–1300 °C, and Nd could be separated as Nd<sub>2</sub>O<sub>3</sub> from silver by oxidation of the Nd–Ag melt in the air [124]. According to Moore et al. [125], after a quick arc or induction melting of the magnet–copper mixture (holding time 1–2 min.), a Cu–Nd rich intermetallic phase was formed upon slow cooling 350 K/min.) on the outer shell of the ingot, when a higher fraction of copper was used in the mixture feed (Cu > 21.5 %, in weight percentage unless otherwise stated). The bath temperature of arc melting could not be detected and controlled, but the induction melting temperature was reported to be in the range of 1450–1500 °C. The Nd–Cu-rich shell (nonmagnetic) is brittle and was mechanically fragmented, thus a mechanical separation from the ferromagnetic core (Fe-rich) could be obtained with magnetic separation. However, the separation is not very efficient: ferromagnetic fraction still contains about 10.4 % Nd and 7.8 % Cu, and nonmagnetic fraction contains 11.4 % Fe

and 43.9 % Cu. Furthermore, further Nd–Cu separation has not been investigated, and the alloy contains almost equal amount of Nd and Cu of 44 %. This could be a fundamental limitation and challenge by using copper as an extractant.

**Molten salt extraction** At Anhui University of Technology (China), Hua et al. [126] reported an integrated process using molten chloride salt (MgCl–KCl) to selectively dissolve Nd and other REEs in the magnet scrap at temperatures of 700–1000 °C with an REE recovery yield of approximately 90 %. NdCl<sub>3</sub> in molten the MgCl<sub>2</sub>–KCl salt mixture can be subsequently electrolyzed to produce a magnesium-neodymium alloy directly. Tanaka et al. [9, 127] proposed a process based on molten fluoride to remove the oxidized rare earths in the NdFeB magnet scrap. In this process, the molten fluoride reacts with the rare-earth oxides (REOs) and the formed REO–fluoride mixture can be used in molten salt electrolysis for REE metal production. The nonoxidized NdFeB scrap does not react, and it obtained as a cleaned master alloy that can be used for the production of new NdFeB magnets. It should be noted that the molten chloride and molten fluoride processes are complementary: molten chloride salts can be used for treatment of nonoxidized NdFeB magnet scrap only, while molten fluoride salts are suitable for the treatment of oxidized NdFeB magnet scrap only. Hua et al. [128] also reported the REE magnet treatment using a fluoride melt, and REEs in NdFeB magnet scrap have been successfully recovered using AlF<sub>3</sub>–NaF melts as the fluorination reagent. Furthermore, active research is going on at TU Delft as part of EU FP7 funded project EREAN (<http://www.erean.eu>) to investigate the salt extraction of NdFeB magnet scrap for further molten salt electro-deposition of the REEs.

**Molten slag extraction** Two different slag treatment methods have been reported. In the glass slag method, the REE alloy is brought into contact with molten B<sub>2</sub>O<sub>3</sub> that is able to selectively oxidize and dissolve the REEs from the NdFeB alloys to form Nd<sub>2</sub>O<sub>3</sub>–B<sub>2</sub>O<sub>3</sub> slag [19]. The slag can be then leached with acid to dissolve Nd and the Nd can be recovered from the leachate by precipitation as Nd(OH)<sub>3</sub>. However, the yield of Nd recovery is relatively poor [129]. The second slag method is the use of CaO–SiO<sub>2</sub>–Al<sub>2</sub>O<sub>3</sub> or CaO–CaF<sub>2</sub> fluxes to extract the REEs in the complex magnet scrap at about 1500 °C [130]. This method is very effective for EOL magnet scrap such as HDD shredder residues with more than 99 % REE recovery. It can separate completely the REEs in the magnet from the ferrous materials in the NdFeB magnet and the mixed scrap. The REE-rich slag can be used for hydrometallurgical extraction, and the separated iron-rich alloy can be a good source of raw materials for steelmaking. The advantages of molten slag extraction are its capability to effectively separate



REEs and iron, and the recovery of both fractions at the same time. According to Tanaka et al. [9], a Japanese group reported an in-plant waste (sludge) separation process where the magnet waste separates into liquid iron and solid rare-earth oxide phases at 1550 °C under argon atmosphere with a low oxygen partial pressure without any flux addition. It was found that all of the REEs in the magnet waste were converted to the oxide phase, and the formed REOs were converted to RE metal through conventional technologies.

**Direct melting** Recently a research group at Shanghai University reported a new method for the treatment of relatively clean NdFeB magnet (without Ni coating) [131]. They first melted the magnet in a vacuum induction furnace at about 1400 °C in a graphite crucible, after which the NdFeB magnet will form REE carbides in the form of NdFeBC alloy. The solidified carbon-saturated alloy containing REE carbides are milled to fine particles and treated with water at room temperature for selective hydrolysis, and the NdC<sub>2</sub> in the alloy phase was converted to Nd(OH)<sub>3</sub>, and the mixed metallic iron and iron carbide are removed by magnet separation. The recovered Nd(OH)<sub>3</sub> is easily calcined to Nd<sub>2</sub>O<sub>3</sub> for direct commercial use or as raw materials for production of Nd metal through molten salt electrolysis. The process of vacuum induction melting (VIM) followed by hydrolysis and magnetic separation (HMS) was named as VIM–HMS method by the authors. This is a very clean process, and no toxic chemicals are used and no significant solid residues or effluent are generated. However, the method was developed with very clean magnets, and it is not clear if the method is suitable for EOL magnet scrap with low rare-earth content and highly complex compositions. Contamination of other transition metals in the NdFeB magnet-bearing streams such as copper and nickel may be a challenge.

In the EU FP7-funded project REEcover (<http://www.reecover.eu>), an alternative pyrometallurgical treatment of a REE-containing ferrous fraction from physically upgraded WEEE shredder products was reported recently [132]. The WEEE shredder product was first thermal demagnetized, and a fine fraction (after screening) was ground and screened to 75 µm [54]. The fraction above 75 µm is used for pyrometallurgical treatment, and the fraction below 75 µm can be treated directly with hydrometallurgical leaching. The upgraded ferrous stream from the shredder product contains about 6500 ppm REEs, originating from NdFeB permanent magnet (Nd, Pr, and Dy). It was melted at 1600 °C under reducing conditions in the graphite crucible for one-hour holding time. A complete separation of REEs from the ferrous fraction was achieved, and all REEs in the scrap were oxidized into a slag phase. The amount of the resulted slag is small, but with a relatively high concentration of REEs (over 33 % in REO form). The REE-

bearing slag contains also Al<sub>2</sub>O<sub>3</sub>, SrO, BaO, SiO<sub>2</sub>, and CaO, with a melting temperature in the range of 1320–1400 °C. The REE-concentrated slag is used for REE separation and extraction through hydrometallurgical leaching and further precipitation to produce more pure REOs, and finally for producing REE metal or alloy through molten salt electrolysis. More research is under way to optimize the melting conditions, e.g., using fluxing agent for enhanced REEs slag formation, to achieve more favorable and selective leaching recovery of REEs. However, the resulting concentration of REEs in the slag will be much lower than the above reported figures.

### *Direct Electrochemical Refining*

Electrochemical processes can be applied to the metallic magnet scrap based on electro-refining principles in the molten salt [9, 133]. In a molten salt electro-refining cell, the impure REE magnet scrap is used as starting anode, and the REEs (Nd and/or Dy) can be selectively dissolved in a molten chloride or fluoride electrolyte, whereas on the cathode, only REEs are preferably deposited forming Nd-based alloys. In the cell, a REE–an iron group metal alloy is used as bipolar electrode (diaphragm).

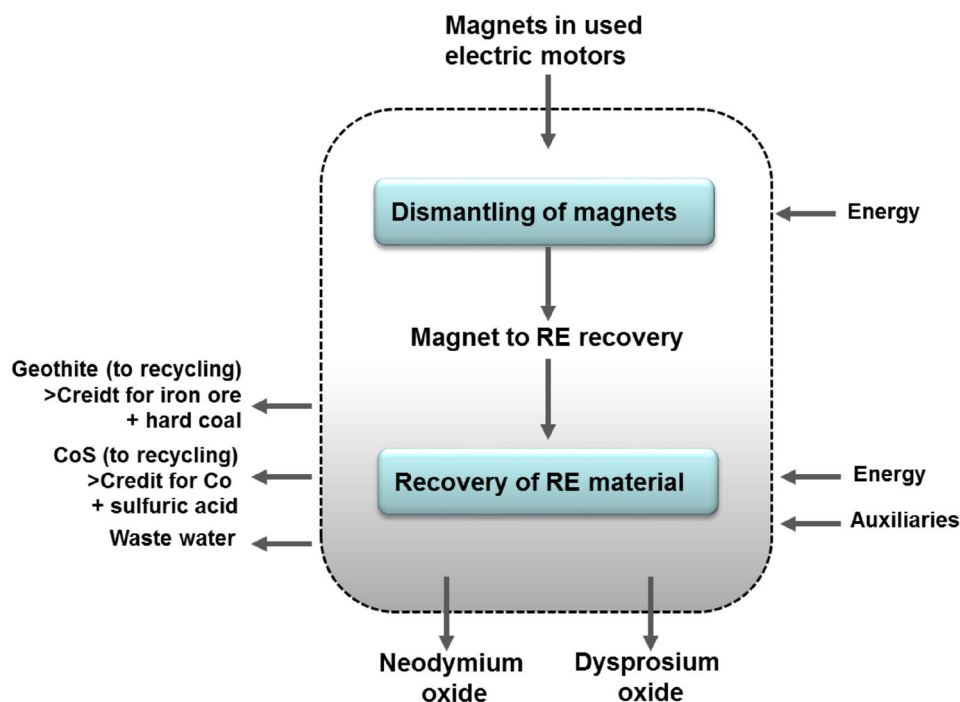
High-temperature processing of NdFeB magnet scrap has also been reviewed by Takeda and Okabe [10], and they described advantages of the so-called dry process over the hydrometallurgical “wet processes.” They predicted that the EOL scrap from products is expected to be generated in the future at a constant volume such as the large magnets in EVs and HEVs. Therefore, efficient mechanisms and processes for recovering Nd and Dy from these types of product scrap should be developed for the future needs. Firdaus et al. [11] indicated a number of challenges for REE recovery from NdFeB magnet scrap: (1) the different mix of wastes produced—the complexity of magnet waste composition; (2) the effects of contaminants on the recycling process; (3) optimization for mutual separation of REEs (Nd, Dy, Pr); and (4) economic feasibility of the recycling and recovery process and lifecycle of the magnet-containing products (i.e., magnet scrap availability).

### **Life Cycle Assessments**

The environmental impact of the life cycle of the REEs used in specific products like magnets can be studied with conventional LCA, the so-called attributional LCA—see, e.g., Finnveden et al. [134]. LCA is a well-developed and acknowledged methodology backed by obligatory ISO Standards 14040 and 14044. The partners of the project MORE, a German national-funded project, addressed the recycling of electric drive motors from the automotive



**Fig. 12** System boundaries of the MORE LCA (recovery of pure neodymium and dysprosium oxides) Adapted from Walachowicz et al. [136]



**Table 5** Generic composition of the NdFeB magnets for the MORE LCA, adapted from Walachowicz et al. [136]

| Elements            | Fe | B | Co | Nd | Dy |
|---------------------|----|---|----|----|----|
| Mass percentage (%) | 67 | 1 | 2  | 20 | 10 |

sector [135]. Within the project, the reuse of NdFeB magnets, reuse of the magnetic alloy as well as REE recovery were investigated. LCA results are available for all these three different routes [136]. In this article, only an excerpt of the LCA results for the raw material route is presented as it was identified as the most promising approach. The MORE LCA was conducted according to ISO 14040/44 including a critical review by an independent external expert and is based on small pilot-scale data. Figure 12 shows the system boundaries of the LCA for the recovery of pure neodymium and dysprosium oxides from magnets used in electric motors via a final solvent-extraction step.

For the different LCAs conducted for the different MORE process routes, the partners agreed upon the generic composition of a typical NdFeB magnet given in Table 5.

The functional unit for this LCA is defined as recovery of 1 kg REE oxide (Nd: Dy ratio = 2:1). It should be highlighted that for the credit calculation for the recovered REE, a mass allocation as well as an economic allocation was used (see data in Table 6).

Figure 13 below demonstrates the LCA results for the recovery of pure Nd<sub>2</sub>O<sub>3</sub> and Dy<sub>2</sub>O<sub>3</sub> compared to the

**Table 6** Comparison of primary Nd and Dy production data\* (GWP). Adapted from Walachowicz et al. [136]

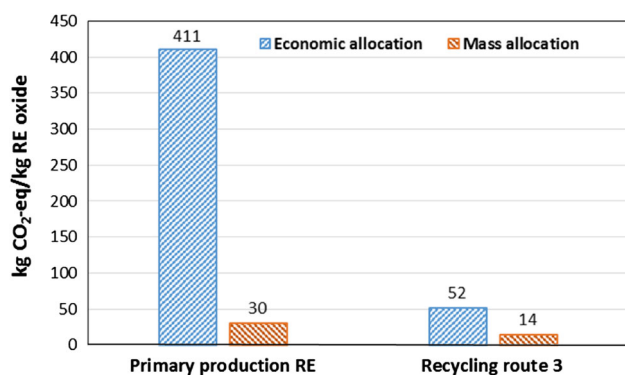
| Allocation approach   | Nd                               | Dy   |
|-----------------------|----------------------------------|------|
|                       | [kg CO <sub>2</sub> -eq./kg REE] |      |
| Economic allocation** | 131                              | 1159 |
| Mass allocation       | 35                               | 35   |

\* PE international data for MORE, 2013

\*\* REE prices in summer 2013

primary production route. The necessary additional step from REE oxide to REE metal was taken into account for this calculation.

Also the results for all other environmental impact categories show net benefits. Therefore, the overall recycling process seems to be very promising from an ecological point of view in comparison to the primary production of REEs. However, the good environmental performance should be verified in future LCA studies if further scale-up takes place especially considering that the collection of comprehensive LCA data for the primary routes of rare earths is a pending and challenging task. Sprecher et al. [137] compared the environmental impact of the primary production process of 1 kg of NdFeB permanent magnets with two alternative recycling processes. This work focused on the recycling of NdFeB magnets from HDDs (hard disk drives). The difference of the two recycling processes is the pretreatment step for the HDDs from EOL computers (manual dismantling of the NdFeB magnets vs.



**Fig. 13** Global warming potential, recovery of pure neodymium and dysprosium oxides (solvent-extraction route) Adapted from Walachowicz et al. [136]

shredding of the HDDs). Again the recycling processes show an improvement over primary production of NdFeB magnets from an environmental point of view. It should be noted that the recycling process with the manual dismantling step shows superior results compared with the alternative which includes shredding of the HDDs. The independent results of the MORE partners [135, 136] and other researchers [137] confirmed the ecological advantages of recycling approaches for NdFeB magnets compared with primary production.

## Concluding Remarks

This review paper addresses the recycling potential of NdFeB permanent magnets and the identified physical and metallurgical separation and recovery technologies. It is indicated that the annual consumption and demand of the REE magnets are continuously increasing, and the recycling of the REEs from the EOL permanent magnets will be complementary to the supply shortage in the long run. REE recovery from the manufacturing scrap and waste (swarf) of the NdFeB magnets is already a good industrial practice with mature technologies.

However, recycling and recovery of REEs (Nd, Pr, Dy, Tb) from EOL magnets are challenging due to their relatively small sizes used in consumer electrical and electronics, as well as in the conventional automotive industry. Shredding without pre-dismantling of the EOL consumer products will pose great difficulties for up-concentration of the permanent magnets. Development of advanced and low-cost of pre-disassembling technologies or equipment will be crucial for the metallurgical recovery of these critical elements. This has been demonstrated by the developments for computer hard disk drives and air conditioners by Hitachi and Mitsubishi, Birmingham University, Delft University of Technology, and more from WEEE streams in a number of active European initiatives such as EREAN

(<http://erean.eu>), REEcover (<http://www.reecover.eu>), REE4EU (<http://www.ree4eu.eu>) consortia and other global government supports in Japan and USA reported in recent years.

There are many different types of metallurgical recovery methods identified from the literature; however, most of them have been studied for relatively clean and highly concentrated new or manufacturing magnet scrap and waste. Adaptation of these technologies to the dilute and complex EOL magnet scrap is very much needed. At the same time, recovery of the major metals and materials in the dilute magnet waste stream (e.g., shredder products) is an important factor for the total success of the permanent magnet and REE recycling. It is believed that no single metallurgical process can be successful for the efficient recovery of the REEs from their EOL secondary resource, and a combination of hydrometallurgical, pyrometallurgical, and/or electrochemical technologies will be the future solution. It is expected that through the globally increasing efforts (Europe, Japan, USA, China) for the research and development, establishment of technologically and economically feasible REE magnet recycling industry could be realized in the near future, if the NdFeB permanent magnet market continue to develop.

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